than any $d_2$ thus far found among the rare gases, and reacts with $3s_5$ (from which it is separated by only 1 $\text{cm}^{-1}$) to produce Paschen-Back effects.*

The very large discrepancy in $2s_2$ is not so easy to account for. It looks, almost, as if the level were improperly classified, although it appears to be in about the correct position. Perhaps $2s_2$ and $3s_1'$ should be interchanged.

The level $4d_5$ seems to have an abnormally low value when compared with the other rare gases, all of which have values about 1.4. Not too much importance can be placed on this point, however, for this configuration is badly distorted. Indeed, $4d_5$ seems to be far below its usual position, being lower than $4d_6$ which is usually the lowest level of the $p^5d$ configuration.

The levels $3p_8$ and $3p_9$ are only 5.5 $\text{cm}^{-1}$ apart and perturb each other sufficiently to distort the patterns of lines involving them. Their $g$ values were determined by methods already described.[9] (See Fig. 2.)

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* The calculations of the Paschen-Back interaction of $3s_5$ and $4d_2$ will be discussed in a separate communication.

[9] J. B. Green and J. F. Eichelberger, Phys. Rev. **56**, 51 (1939); J. B. Green and J. A. Peoples, *ibid*. **56**, 54 (1939).

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DECEMBER 15, 1940 PHYSICAL REVIEW VOLUME 58

# Field Dependence of the Intrinsic Domain Magnetization of a Ferromagnet

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(Received July 31, 1940)

In this paper, the variation of the intrinsic domain magnetization of a ferromagnetic with the external magnetic field, is obtained. The basis of the treatment is the exchange interaction model amplified by explicit consideration of the dipole-dipole interaction between the atomic magnets. Approximations appropriate to low temperatures and equivalent to those used by Bloch in his derivation of the $T^{3/2}$ law, are introduced. The resultant expression for the intrinsic volume susceptibility decreases slowly with increasing field; at high fields the functional dependence is as the inverse square root of the field. The variation with temperature is linear; at room temperature and for fields of about 4000 gauss, the order of magnitude of the (volume) susceptibility is $10^{-4}$. The results are compared with experiment and satisfactory agreement is found.

## I. Introduction

CLOSELY allied to the problem of the temperature variation of the intrinsic magnetization, **M**, of a ferromagnetic body, is the problem of its variation with an external magnetic field, **H**. This intrinsic magnetization is characteristic of a single ferromagnetic domain and is identical with the experimentally observed magnetization when "technical saturation" has been achieved, i.e., when all of the domains in all of the individual crystal grains of the specimen have parallel magnetization vectors.[1]

When technical saturation has been reached, there are still, as a result of temperature agitation, some atomic magnetic moments which are not oriented in the direction of **H**. Further increase of magnetization is then to be ascribed to the progressive alignment of the temperature disoriented atomic magnets by an increasing field. The phenomenon is physically similar to that which obtains in the magnetization of a paramagnetic substance; however, the existence of the strong ferromagnetic coupling forces between the atomic magnets changes completely[2] both the magnitude of the effect, and its de-

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[1] "Technical saturation" is achieved at fields of 2000–4000 gauss, depending on the metallurgical treatment of the (polycrystalline) specimen.

[2] Cf. Eq. (30), (31) below.

pendence upon the external magnetic field and upon the temperature.

The problem of the variation of **M** with **H** has been treated by Akulov[3] on the basis of the phenomenological Weiss-Heisenberg internal field theory;[4] his result is in strong disagreement with subsequent experiments.[5]

The basis of the treatment of the variation of **M** with **H** given in the present paper is the exchange interaction model of a ferromagnetic,[6] wherein the electrons responsible for the ferromagnetism are anchored to the atomic cores and do not wander about the metallic interior. The essential approximation used in the treatment of this model is that the condition of "quasi-saturation" obtains; *viz.*, the percent deviation of the magnetization $\mathbf{M}(T, H)$ from its maximum possible value, $\mathbf{M}_0$, is small. This condition always prevails at temperatures sufficiently below the Curie temperature.[7]

It has also been found possible to take account of the magnetic interactions between the atomic magnets. These interactions are important, since, in most of the experiments thus far performed, the magnetic energy of an atom due to the magnetic moments of all of the other atoms, $\leqq 4\pi\beta M_0/3$ is usually larger than the magnetic energy due to an external magnetic field, $\leqq \beta H$.[8] It will be seen that the effect of the magnetic interactions cannot be expressed correctly by replacement of $H$ by $H+\gamma M$ [9] in the final expression for the magnetization (cf. (30), (31), (32), below).

## II. The Hamiltonian

The exchange interaction model of a ferromagnetic, amplified by the inclusion of magnetic interactions, is described, in the space of the atomic spin variables by the following Hamiltonian,[9a]

$$\mathrm{H} = -\tfrac{1}{2}\sum_{l,m=1}^{N} 2J_{lm}(R_{lm})\mathbf{S}_l\cdot\mathbf{S}_m + \tfrac{1}{2}\sum_{l,m=1}^{N}(4\beta^2/R^5{}_{lm})(R^2{}_{lm}\mathbf{S}_l\cdot\mathbf{S}_m - 3\mathbf{S}_l\cdot\mathbf{R}_{lm}\mathbf{S}_m\cdot\mathbf{R}_{lm}) - \sum_{l=1}^{N} 2\beta S_l{}^{(z)}H. \quad (1)$$

Here, $N$ is the total number of atoms, while the sums over $l$ and $m$ each run from 1 to $N$, summands with $l=m$ being omitted. $R_{lm} = |\mathbf{R}_l - \mathbf{R}_m|$ is the distance between the centers of gravity of the $l$th and $n$th atoms; $J_{lm}$ the exchange integral between these atoms; $\mathbf{S}_l$ the spin angular momentum operator of the atom at $\mathbf{R}_l$ in units of $\hbar$; the magnetic field $H$ is directed along the $z$ axis.

The first term in H is the Heisenberg exchange energy expressed in terms of the atomic spin operators (Dirac vector model).[10] The second term arises from the magnetic dipole-dipole interaction between electrons on different atoms. The effects of exchange and of higher magnetic poles on the magnetic interaction are both neglected;[11] hence, the centers of gravity of the atoms constitute the

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[3] N. S. Akulov, Zeits. f. Physik **69**, 822 (1931). See, also, R. Becker and W. Döring, *Ferromagnetismus* (Julius Springer, Berlin, 1939), p. 35.

[4] See, for example, J. H. Van Vleck, *Theory of Electric and Magnetic Susceptibilities* (Oxford University Press, 1932), Ch. 12.

[5] A full discussion of the various experimental results and comparison with theory is given below in Section VI.

[6] W. Heisenberg, Zeits. f. Physik **49**, 619 (1928).

[7] The exchange interaction model, with the specimen in the condition of quasi-saturation, has been approximately treated by Bloch in his derivation of the temperature variation of the intrinsic magnetization. Cf. Zeits. f. Physik **61**, 206 (1930); **74**, 295 (1932). Using mathematical methods different from those in the present paper, Bloch derives the well-known $T^{\frac{3}{2}}$ law for atoms with spin $S=\frac{1}{2}$. Bloch's methods have been extended by Möller to the case $S>\frac{1}{2}$. Cf. Zeits. f. Physik **82**, 559 (1933).

[8] $\beta$ is the Bohr magneton.

[9] $\gamma$ is a numerical factor whose value lies between 0 and $4\pi$.

[9a] It is to be noted that $(\mathbf{S}_l)^2$ commutes with the Hamiltonian of Eq. (1); therefore, the magnitude of the spin of each atom has a definite integral or half-integral value, $S$, and thus, each atomic magnetic moment should be an integral multiple of the Bohr magneton. However, experimental values of the intrinsic magnetization extrapolated to absolute zero, i.e. experimental values of $M_0$, give for the atomic magnetic moments, Fe:$2.2\beta$; Ni:$0.6\beta$; Co:$1.7\beta$. The deviations of these magnetic moments from integral multiples of $\beta$ is due, presumably, to the interchange of electrons between the atomic $3d$ shells, where the electrons contribute to the ferromagnetism, and the conduction states, where they do not. The problem of the extension of the exchange interaction model to include these interchanges has not yet been solved.

[10] See reference 4, Ch. 12.

[11] Magnetic quadrupole interactions have been invoked to explain ferromagnetic anisotropy, cf. R. Becker, *Theorie der Electrizität*, Vol. II (Teubner, Leipzig, 1933) pp. 177–183; in order that these, alone, account for the observed ansiotropy their ratio to the dipole interactions need only be $\leqq 1/10$. In this connection it should further be mentioned that the use of magnetic quadrupole interactions to explain ferromagnetic anisotropy has been criticized by Van Vleck, Phys. Rev. **52**, 1178 (1937), p. 1188.

locations of magnetic dipoles, each with moment $2\beta S$. The last term is the interaction of the magnetic moment of each atom with the external magnetic field.

It is to be noted that spin orbit effects have been neglected in Eq. (1). How these effects are to be treated and what influence they have on the final result will be discussed in a later section.

## III. EIGENVALUES OF THE HAMILTONIAN

The first step in finding the magnetization is the determination of the eigenvalues, E of H. In conformance with the essential approximation of "quasi-saturation," $[M_0 - M(T, H)]/M_0 \ll 1$ mentioned in the introduction, only those eigenvalues, E, are sought whose corresponding eigenfunctions, $\Psi_{\mathrm{E}}$, specify states where the expectation value of the $z$ component of the total spin of the specimen

$$\sum_{l=1}^{N} S^{(z)}{}_l$$

is close to its maximum possible value $NS$.[12]

To proceed, it is convenient to introduce the operators:

$$S^+{}_l = S^{(x)}{}_l + iS^{(y)}{}_l, \quad S^-{}_l = S^{(x)}{}_l - iS^{(y)}{}_l, \quad \mathfrak{n}_l = S - S^{(z)}{}_l. \tag{2}$$

$n_l$ the eigenvalue of $\mathfrak{n}_l$, will be called the "spin deviation" of the $l$th atom; for the state

$$\Psi_{n_1, \cdots, n_l, \cdots n_N} \equiv \Psi_{n_l}$$

which is an eigenstate of $\mathfrak{n}_l$ and $S^{(z)}{}_l$, $n_l$ obviously represents the difference between the $z$ component of the spin of the $l$th atom and its maximum value. Also, by way of notation $\langle \mathfrak{n}_l \rangle_{\mathrm{E}}$ is the expectation value of the spin-deviation operator, $\mathfrak{n}_l$, averaged over the eigenstate

$$\Psi_{\mathrm{E}} = \sum_{n_1, \cdots, n_N} b_{\mathrm{E}}(n_1, \cdots, n_l, \cdots n_N) \Psi_{n_1, \cdots, n_l, \cdots n_N},$$

i.e.,

$$\langle \mathfrak{n}_l \rangle_{\mathrm{E}} = \sum_{n_1, \cdots, n_N} |b_{\mathrm{E}}(n_1, \cdots, n_l, \cdots, n_N)|^2 n_l,$$

whereas,

$$\langle\langle \mathfrak{n}_l \rangle\rangle_{\mathrm{Av}} \equiv \textstyle\sum_{\mathrm{E}} \langle \mathfrak{n}_l \rangle_{\mathrm{E}} e^{-\mathrm{E}/kT} / \sum_{\mathrm{E}} e^{-\mathrm{E}/kT}$$

is the expectation value of the spin-deviation operator when the temperature of the specimen is $T$, and involves, first an average over $\Psi_{\mathrm{E}}$, and then an average over the Boltzmann distribution of the eigenstates of the specimen.

The operators of (2) have the following properties,

$$\begin{aligned} S^+{}_l \Psi_{n_l} &= (2S)^{\frac{1}{2}} (1 - (n_l - 1)/2S)^{\frac{1}{2}} (n_l)^{\frac{1}{2}} \Psi_{n_l - 1}, \\ S^-{}_l \Psi_{n_l} &= (2S)^{\frac{1}{2}} (n_l + 1)^{\frac{1}{2}} (1 - n_l/2S)^{\frac{1}{2}} \Psi_{n_l + 1}, \quad \mathfrak{n}_l \Psi_{n_l} = n_l \Psi_{n_l}. \end{aligned} \tag{3}$$

Introducing the well-known creation and destruction operators defined by[13]

$$a^*{}_l \Psi_{n_l} = (n_l + 1)^{\frac{1}{2}} \Psi_{n_l + 1}, \quad a_l \Psi_{n_l} = (n_l)^{\frac{1}{2}} \Psi_{n_l - 1}, \tag{4}$$

one obtains, upon comparing (3) and (4)

$$S^+{}_l = (2S)^{\frac{1}{2}} (1 - a^*{}_l a_l / 2S)^{\frac{1}{2}} a_l, \quad S^-{}_l = (2S)^{\frac{1}{2}} a^*{}_l (1 - a^*{}_l a_l / 2S)^{\frac{1}{2}}, \quad \mathfrak{n}_l = a^*{}_l a_l. \tag{5}$$

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[12] It is to be noted that $\sum_{l=1}^{N} S^{(z)}{}_l$ does not commute with the magnetic interaction portion of the Hamiltonian (1).

[13] In Eq. (4), $n_l$ is allowed to run from 0 to $\infty$ rather than from 0 to $2S$ as in Eq. (3). The discrepancy is only apparent, since the transition from states with $n_l \leqq 2S$ to states with $n_l > 2S$ will never occur. e.g.

$$S^-{}_l \Psi_{2S} = (2S)^{\frac{1}{2}} (2S + 1)^{\frac{1}{2}} (1 - 2S/2S)^{\frac{1}{2}} \Psi_{2S+1} = 0.$$

When substituted into the Hamiltonian of Eq. (1), (2) and (5) give:

$$\begin{aligned}
\mathrm{H}=&-\sum_{l,m} J_{lm}(2Sa^*{}_l(1-a^*{}_la_l/2S)^{\frac12}(1-a^*{}_ma_m/2S)^{\frac12}a_m+S^2-2Sa^*{}_la_l+a^*{}_la_la^*{}_ma_m)\\
&+\tfrac12\sum_{l,m}(4\beta^2/R^3{}_{lm})(2Sa^*{}_l(1-a^*{}_la_l/2S)^{\frac12}(1-a^*{}_ma_m/2S)^{\frac12}a_m+S^2-2Sa^*{}_la_l+a^*{}_la_la^*{}_ma_m)\\
&+\tfrac12\sum_{l,m}(4\beta^2/R^3{}_{lm})(-3)([z^2{}_{lm}(S^2-2Sa^*{}_la_l+a^*{}_la_la^*{}_ma_m)]\\
&+[z_{lm}\overset{+}{R}_{lm}(S-a^*{}_la_l)(2S)^{\frac12}a^*{}_m(1-a^*{}_ma_m/2S)^{\frac12}+z_{lm}\overset{-}{R}_{lm}(S-a^*{}_la_l)(2S)^{\frac12}(1-a^*{}_ma_m/2S)^{\frac12}a_m]\\
&+(2S/4)[(\overset{+}{R}_{lm})^2a^*{}_l(1-a^*{}_la_l/2S)^{\frac12}a^*{}_m(1-a^*{}_ma_m/2S)^{\frac12}+(\overset{-}{R}_{lm})^2(1-a^*{}_la_l/2S)^{\frac12}\\
&\times a_l(1-a^*{}_ma_m/2S)^{\frac12}a_m+2\overset{-}{R}_{lm}\overset{+}{R}_{lm}a^*{}_l(1-a^*{}_la_l/2S)^{\frac12}(1-a^*{}_ma_m/2S)^{\frac12}a_m])\\
&-\sum_{l=1}^{N}2\beta H(S-a^*{}_la_l),
\end{aligned}\tag{6}$$

with $\overset{+}{R}_{lm}=x_{lm}+iy_{lm};\quad \overset{-}{R}_{lm}=x_{lm}-iy_{lm}.$

The condition of quasi-saturation

$$1\gg\tfrac12[M_0-M(T,H)]/M_0=\langle\langle\mathfrak{n}_l\rangle\rangle_{\mathrm{Av}}/2S=\langle\langle a^*{}_la_l\rangle\rangle_{\mathrm{Av}}/2S$$

is now invoked to effect the following approximations in H.

I. Replacement of $(1-a_l{}^*a_l/2S)^{\frac12}$ by 1. Such a replacement appears reasonable since the expectation value of $(1-a^*{}_la_l/2S)^{\frac12}$ is $\cong(1-\langle\langle\mathfrak{n}_l\rangle\rangle_{\mathrm{Av}}/2S)^{\frac12}\cong1$.

II. Neglect of terms proportional to $a^*{}_la_la^*{}_ma_m=\mathfrak{n}_l\mathfrak{n}_m$. Assuming that there is no correlation in the location of the different spin deviations, one obtains for the expectation value of these terms, $\langle\langle\mathfrak{n}_l\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}\cong\langle\langle\mathfrak{n}_l\rangle\rangle_{\mathrm{Av}}\langle\langle\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}$ which is smaller than the expectation value of $2Sa^*{}_la_l=2S\langle\langle\mathfrak{n}_l\rangle\rangle_{\mathrm{Av}}$, by a factor $\langle\langle\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}/2S\ll1$.

III. Neglect of terms proportional to $(2S)^{\frac12}a^*{}_la_la^*{}_m=(2S)^{\frac12}\mathfrak{n}_la^*{}_m$. These are terms which cause the system to make transitions between states of different total spin. Unlike terms of the type $2Sa^*{}_la^*{}_m$ which also cause such transitions, they are different from zero only for transitions taking place near atoms on which spin deviations are already present; the ratio of the number of transitions arising from the two types of terms is thus $\cong\langle\langle\mathfrak{n}_l\rangle\rangle_{\mathrm{Av}}(2S)^{-\frac12}\ll1$.

A further discussion of the nature of the approximations I, II, III, is given in Appendix I. In anticipation of results to be obtained below (cf. Eq. (15′) and following text), it may be pointed out that, if the magnetic dipole-dipole interactions are omitted, these approximations lead to the same energy levels as obtained by Bloch in his derivation of the $T^{\frac32}$ law,[7] and are thus equivalent to the approximations of his method.

An additional simplification of the Hamiltonian results from the fact that summations of the form:[14]

$$\textstyle\sum_{l,m}f(R_{lm})x_{lm}z_{lm}a^*{}_m=\sum_m a^*{}_m\sum_h f(R_h)x_hz_h$$

are zero. This fact is obvious if the direction of **H**, i.e., the $z$ axis, coincides with one of the crystallographic cubical axes. In the more general case, a transformation of the sum over $h$ to Cartesian coordinates which are referred to the cubical axes readily leads to the same result.

Applying approximations I, II, III, and the theorem just stated, one obtains for the Hamiltonian (6)

$$\begin{aligned}
\mathrm{H}=&\,C-\textstyle\sum_{l,m}2SJ_{lm}(a^*{}_la_m-a^*{}_la_l)+\frac12\sum_{l,m}(4\beta^2/R^3{}_{lm})(2S)(a^*{}_la_m-a^*{}_la_l)\\
&+\tfrac12\textstyle\sum_{l,m}(4\beta^2/R^3{}_{lm})(-3)(2S)(\frac12[x^2{}_{lm}+y^2{}_{lm}]a^*{}_la_m-z^2{}_{lm}a^*{}_la_l+\frac14[(\overset{+}{R}_{lm})^2a^*{}_la^*{}_m+(\overset{-}{R}_{lm})^2a_la_m])\\
&+\textstyle\sum_l 2\beta Ha^*{}_la_l,
\end{aligned}\tag{7}$$

with $C=-\sum_{l,m}J_{lm}S^2-2\beta SNH-\frac12\sum_l2\beta S\{\sum_m(-2\beta S)/R^3{}_{lm}(1-3z^2{}_{lm}/R^2{}_{lm})\}.$

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[14] $f(\mathbf{R}_{lm})$ is any function of $\mathbf{R}_{lm}$ and $\mathbf{R}_h\equiv\mathbf{R}_l-\mathbf{R}_m$. The two sums are identical provided that contributions coming from atoms on the surface of the specimen are neglected; the justification of this neglect is given in the text before Eq. (11).

The numerical constant $C$ is the value of the energy when all of the atomic magnets point in the direction of the field $\mathbf{H}$. The first sum in $C$ is the exchange energy, the second the interaction between the atomic magnets and the external field, and the last the mutual dipole-dipole interaction of the atomic magnets. (In fact, the expression in the curly brackets is the sum of the Lorentz local-field, $4\pi M_0/3$, and the demagnetizing field, as will be shown in another connection below. Cf. text after Eqs. (44), (45) in Appendix II.

From the form of the Hamiltonian (7), it is apparent that the "spin deviations," specified by the integers $n_l$ are not localized on any one atom, but are "propagated" through the crystal. That this propagation is essentially of a wave-like character is seen by the introduction of new variables defined by the following relations:

$$\begin{aligned} a_\lambda &= N^{-\frac{1}{2}} \textstyle\sum_l \exp\,(i\mathbf{K}_\lambda\cdot\mathbf{R}_l)a_l\,; & a_l &= N^{-\frac{1}{2}} \textstyle\sum_\lambda \exp\,(-i\mathbf{K}_\lambda\cdot\mathbf{R}_l)a_\lambda, \\ a^*_\lambda &= N^{-\frac{1}{2}} \textstyle\sum_l \exp\,(-i\mathbf{K}_\lambda\cdot\mathbf{R}_l)a_l\,; & a^*_l &= N^{-\frac{1}{2}} \textstyle\sum_\lambda \exp\,(i\mathbf{K}_\lambda\cdot\mathbf{R}_l)a^*_\lambda. \end{aligned} \tag{8}$$

The $a_\lambda$ and $a^*_\mu$ satisfy the relation:

$$a_\lambda a^*_\mu - a^*_\mu a_\lambda = \delta_{\mu\lambda}, \tag{9}$$

since from Eq. (4),

$$a_l a^*_m - a^*_m a_l = \delta_{ml}. \tag{10}$$

In Eq. (8), $\mathbf{R}_l$ is the vector from an arbitrary origin to the $l$th atom, whose magnitude measures the corresponding distance in units of the lattice constant, in contrast to the formulae of the above text where $|\mathbf{R}_l|$ gives distance in centimeters. $\mathbf{K}_\lambda$ is a reduced wave-vector; the usual periodicity conditions require its components to take the values

$$\mathrm{K}^{(x)}{}_\lambda = 2\pi\lambda_x/G_x, \quad \mathrm{K}^{(y)}{}_\lambda = 2\pi\lambda_y/G_y, \quad \mathrm{K}^{(z)}{}_\lambda = 2\pi\lambda_z/G_z,$$

where $\lambda_x$, $\lambda_y$, $\lambda_z$ assume any integral values between $-\frac{1}{2}G_x$ and $\frac{1}{2}G_x-1$, $-\frac{1}{2}G_y$ and $\frac{1}{2}G_y-1$, $-\frac{1}{2}G_z$ and $\frac{1}{2}G_z-1$, respectively.[15]

The replacement of the $a_l$ by the $a_\lambda$ constitutes the first step in the evaluation of the eigenvalues of the Hamiltonian. To carry out this replacement, one has to evaluate sums of the type,

$$\textstyle\sum_{l,m} f(\mathbf{R}_{lm})a^*_l a_m.$$

Upon application of (10), this sum becomes

$$\sum_{l,m,\lambda,\lambda'} N^{-1}f(\mathbf{R}_{lm})a^*_{\lambda'}a_\lambda \exp\,\{i(\mathbf{K}_{\lambda'}\cdot\mathbf{R}_l - \mathbf{K}_\lambda\cdot\mathbf{R}_m)\}.$$

Introducing the vector $\mathbf{R}_h = \mathbf{R}_l - \mathbf{R}_m = \mathbf{R}_{lm}$ one obtains

$$\sum_{l,h,\lambda,\lambda'} N^{-1}f(\mathbf{R}_h)a^*_{\lambda'}a_\lambda \exp\,\{i(\mathbf{K}_{\lambda'} - \mathbf{K}_\lambda)\cdot\mathbf{R}_l)\}\exp\,\{i\mathbf{K}_\lambda\cdot\mathbf{R}_h\}.$$

Now, the summations over $l$ and $h$ can be carried out independently if one neglects contributions arising from the surface of the specimen. Such contributions are obviously negligible for the exchange forces. In this case, the summation over $h$ comprises only nearest neighbors of $l$, and hence surface terms arise only if the atom $l$ is, itself, on the surface. However, the number of such terms is smaller than the total number of terms by a factor $\leqq 1/G$. As far as the magnetic forces are concerned, although their short range character is not immediately obvious, one can show that, in the sum over $h$, again only small values of $|\mathbf{R}_h|$ are important; hence the argument given just above again applies. The summation over $l$ then yields the factor $N\delta_{\lambda'\lambda}$ and the quadruple sum reduces to

$$\textstyle\sum_\lambda(\sum_h f(\mathbf{R}_h)\exp\,\{i\mathbf{K}_\lambda\cdot\mathbf{R}_h\})a^*_\lambda a_\lambda.$$

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[15] $G_x$, $G_y$, $G_z$ are the lengths of the specimen in the $x$, $y$, $z$, directions, divided by the lattice spacing.

Treating all of the summations in H by this method, one obtains:

$$\mathrm{H}=C+\sum_\lambda A_\lambda a^*{}_\lambda a_\lambda+\sum_\lambda(\tfrac{1}{2}B_\lambda a_\lambda a_{-\lambda}+\tfrac{1}{2}B^*{}_\lambda a^*{}_\lambda a^*{}_{-\lambda}), \tag{11}$$

where

$$A_\lambda=3\beta\sum_h(-2\beta S/a^3R^3{}_h)(1-3z^2{}_h/R^2{}_h)+\beta\sum_h(2\beta S/a^3R^3{}_h)(1-3z^2{}_h/R^2{}_h)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))$$
$$+\sum_h 2SJ_h(R_h)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))+2\beta H. \tag{12}$$

$$B_\lambda=-3\beta\sum_h(2\beta S/a^3R^3{}_h)(x^2{}_h-y^2{}_h-2ix_hy_h)/R^2{}_h$$
$$+3\beta\sum_h(2\beta S/a^3R^3{}_h)((x^2{}_h-y^2{}_h-2ix_hy_h)/R^2{}_h)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h)), \tag{13}$$

$a$ being the lattice distance.

The sums over $h$ in $A_\lambda$ and $B_\lambda$ are evaluated in Appendix II; the results for $|\mathbf{K}_\lambda|\ll 1$ are:

$$A_\lambda=A_{-\lambda}=2SJ\mathrm{K}_\lambda{}^2+2\beta H+4\pi\beta M_0\sin^2\theta_\lambda, \tag{14}$$

$$B_\lambda=B_{-\lambda}=4\pi\beta M_0\sin^2\theta_\lambda e^{-2i\phi_\lambda}. \tag{15}$$

Here, $J\equiv J_h(R_h)$ when $R_h$ is the distance between nearest neighbors; $\theta_\lambda$, $\phi_\lambda$ are polar angles of $\mathbf{K}_\lambda$ with polar axis parallel to the field, **H**. As will be shown later in the text, the values of $A_\lambda$ and $B_\lambda$ which contribute importantly to the magnetization **M** are those with $|\mathbf{K}_\lambda|\ll 1$.

It might be instructive to point out that, if the magnetic interactions are omitted, H reduces to

$$-N\sum_h J_hS^2-2\beta SNH+\sum_\lambda(\sum_h J_h2S(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))+2\beta H)a^*{}_\lambda a_\lambda; \tag{15'}$$

since the eigenvalues, $n_\lambda$, of $\mathfrak{n}_\lambda\equiv a^*{}_\lambda a_\lambda$ are 0, 1, 2, 3, $\cdots$ [cf. Eq. (9)], it is seen that the eigenvalues of this Hamiltonian are just the energy values which have been found by Bloch and Möller.[7] In this connection, it is also apparent that $n_\lambda$ can be interpreted as the number of "spin-wave quanta" associated with the wave vector $\mathbf{K}_\lambda$. Further, $\sum_\lambda\langle\langle\mathfrak{n}_\lambda\rangle\rangle_{\mathrm{Av}}$ which is equal to $\sum_l\langle\langle\mathfrak{n}_l\rangle\rangle_{\mathrm{Av}}$, gives directly the expectation value of the deviation from its maximum value of the $z$ component of the total spin of the specimen.

However, the existence of the magnetic interactions, in particular the terms of (11) in $a_\lambda a_{-\lambda}$ and $a^*{}_\lambda a^*{}_{-\lambda}$ which do not commute with $\mathfrak{n}_\lambda$, necessitates further transformation before the Hamiltonian is brought to diagonal form. These transformations are, in order:

$$a_\lambda=2^{-\frac{1}{2}}e^{i\phi_\lambda}(b_\lambda+b_{-\lambda})\,;\quad a_{-\lambda}=2^{-\frac{1}{2}}e^{i\phi_{-\lambda}}(b_\lambda-b_{-\lambda}) \tag{16}$$

and similarly for $a^*{}_\lambda$, $a^*{}_{-\lambda}$. These transformations are defined only for the half-space of $\mathbf{K}_\lambda$, i.e., $-\pi<\mathrm{K}^{(x)}{}_\lambda<\pi$, $-\pi<\mathrm{K}^{(y)}{}_\lambda<\pi$, $0<\mathrm{K}^{(z)}{}_\lambda<\pi$.

$$\begin{aligned} b_\lambda&=l_1c_\lambda+l_2c^*{}_\lambda\,; & b_{-\lambda}&=l_1c_{-\lambda}-l_2c^*{}_{-\lambda}\\ b^*{}_\lambda&=l_1c^*{}_\lambda+l_2c_\lambda\,; & b^*{}_{-\lambda}&=l_1c^*{}_{-\lambda}-l_2c_{-\lambda}\end{aligned} \tag{17}$$

with

$$l_1=\left(\frac{1}{2}\,\frac{A_\lambda+(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}}{(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}}\right)^{\frac{1}{2}};\quad l_2=\left(\frac{1}{2}\,\frac{A_\lambda-(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}}{(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}}\right)^{\frac{1}{2}}. \tag{18}$$

In terms of $c_\lambda$, $c^*{}_\lambda$, the Hamiltonian (11), becomes:

$$\mathrm{H}=C+\sum_\lambda((A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}C^*{}_\lambda C_\lambda+\tfrac{1}{2}(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}-\tfrac{1}{2}A_\lambda). \tag{19}$$

In (19) the sum over $\lambda$ goes over the *whole* of $\mathbf{K}_\lambda$ space, as in (11).

The eigenvalues, E, of H are now immediately available since, by (9), (16), (17), the eigenvalues, $\mathrm{N}_\lambda$ of $C^*{}_\lambda C_\lambda$ are 0, 1, 2, 3, $\cdots$. Thus

$$\mathrm{E}\equiv\mathrm{E}_{\mathrm{N}_\lambda}=C+\sum_\lambda(\tfrac{1}{2}(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}-\tfrac{1}{2}A_\lambda)+\sum_\lambda(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}\mathrm{N}_\lambda. \tag{20}$$

## IV. CALCULATION OF THE MAGNETIZATION, $M(T, H)$

The magnetization is obtained from the partition function, $Z$, by the relation,

$$M=\frac{kT}{V}\frac{\partial}{\partial H}\log Z \tag{21}$$

with

$$\begin{aligned} Z&=\textstyle\sum_{\mathrm{E}}e^{-\mathrm{E}/kT}=e^{-C/kT}\exp\left[-\sum_\lambda\frac{1}{2}((A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}-A_\lambda)/kT\right]\sum_{\mathrm{N}_\lambda}\exp\left[-\sum_\lambda(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}\mathrm{N}_\lambda/kT\right]\\ &=e^{-C/kT}\exp\left[-\textstyle\sum_\lambda\frac{1}{2}((A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}-A_\lambda)/kT\right]\prod_\lambda(1-\exp\left[(A^2{}_\lambda-|B_\lambda|^2)^{\frac{1}{2}}/kT\right])^{-1}. \end{aligned} \tag{22}$$

Thus by (21), (22), and replacing $\sum_\lambda f(\mathbf{K}_\lambda)$ by $G_xG_yG_z(2\pi)^{-3}\int f(\mathbf{K})d\mathbf{K}$, one obtains

$$\begin{aligned} M(T,H)=\frac{2\beta SN}{V}-\frac{G_xG_yG_z2\beta}{(2\pi)^3V}\int\left[\left(\frac{A(\mathbf{K})}{((A(\mathbf{K}))^2-|B(\mathbf{K})|^2)^{\frac{1}{2}}}\right)\left(\frac{1}{\exp\left[((A(\mathbf{K}))^2-|B(\mathbf{K})|^2)^{\frac{1}{2}}/kT\right]-1}\right)\right]d\mathbf{K}\\ -\frac{G_xG_yG_z\beta}{(2\pi)^3V}\int\left[\frac{A(\mathbf{K})}{((A(\mathbf{K}))^2-|B(\mathbf{K})|^2)^{\frac{1}{2}}}-1\right]d\mathbf{K}\equiv M_0-M_T(T,H)-M_\beta(H). \end{aligned} \tag{23}$$

$M_0$ is the value of the magnetization when all of the spins are parallel to each other—complete saturation. $M_T$ is the deviation of $M$ from $M_0$ due to temperature agitation; its magnitude, however, is also dependent on $H$ and the magnetic dipole-dipole interaction. Finally, $M_\beta$ is the deviation of $M$ from $M_0$ due to the dipole-dipole interaction, itself; the magnitude of $M_\beta$ is, in addition, a function of $H$. The presence of the term $M_\beta$ indicates that, even at $T=0$, *complete* saturation can be attained only by the application of a field $H\gg4\pi M_0$.

The $T^{\frac{3}{2}}$ law of Bloch is obtained from (23) by neglecting the magnetic interactions and by setting $\mathbf{H}=0$. Then, from (14) and (15), $A(\mathbf{K})=A(-\mathbf{K})=2SJ\mathrm{K}^2$; $B(\mathbf{K})=B(-\mathbf{K})=0$ and

$$\begin{aligned} M(T)^{\mathrm{Bloch}}\equiv M_0-M_T{}^{\mathrm{Bloch}}&=M_0-G_xG_yG_z2\beta(2\pi)^{-3}V^{-1}\int(\exp\left[2SJ\mathrm{K}^2/kT\right]-1)^{-1}d\mathbf{K}\\ &=M_0(1-\left[M^{-1}{}_0G_xG_yG_zV^{-1}2\beta(2\pi)^{-3}(k/2SJ)^{\frac{3}{2}}(2\pi)(2)(1.3)\right]T^{\frac{3}{2}})\\ &=M_0(1-(\mathrm{const.})T^{\frac{3}{2}}). \end{aligned} \tag{24}$$

It is to be noted that in (24) the integration is extended over all $\mathbf{K}$-space rather than over a cube of side $2\pi$, and the exchange energy $\sum_h2SJ_h(R_h)(1-\exp\left[i\mathbf{K}\cdot\mathbf{R}_h\right])$ is approximated by $2SJ\mathrm{K}^2$. This procedure is permissible provided that $kT/2SJ\ll1$, since in this case the main contributions to the integral come from regions of $\mathbf{K}$-space for which $|\mathbf{K}|\lesseqq(kT/2SJ)^{\frac{1}{2}}$.[16]

Turning now to the evaluation of the integral for $M_T$ in (23), one proceeds most conveniently by finding the difference between $M_T$ and the known integral for $M_T{}^{\mathrm{Bloch}}$ in (24). Thus,

$$M_T-M_T{}^{\mathrm{Bloch}}=\frac{G_xG_yG_z}{(2\pi)^3}\frac{2\beta}{V}\int\left[\left(\frac{A}{(A^2-|B|^2)^{\frac{1}{2}}}\right)\left(\frac{1}{\exp\left[(A^2-|B|^2)^{\frac{1}{2}}/kT\right]-1}\right)-\frac{1}{\exp\left[2SJ\mathrm{K}^2/kT\right]-1}\right]d\mathbf{K} \tag{25}$$

with $A$ and $B$ satisfying relations (14) and (15), *viz.:*

$$A(\mathbf{K})=2SJ\mathrm{K}^2+4\pi\beta M_0\sin^2\theta_{\mathrm{K}}+2\beta H,\quad |B(\mathbf{K})|=4\pi\beta M_0\sin^2\theta_{\mathrm{K}}.$$

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[16] The condition $kT/2SJ\ll1$ is, in practice, not as well fulfilled as is theoretically desirable, e.g. for iron at room temperature $kT/2SJ\lesseqq0.3$.

The important contributions to the integrand of (25) come from regions of $\mathbf{K}$-space for which $|\mathbf{K}|\lesseqqgtr[(4\pi\beta M_0+2\beta H)/2SJ]^{\frac{1}{2}}$. Further, in all practical cases, $(4\pi\beta M_0+2\beta H)/kT\leqq 2\times 10^{-3}$. Thus, in the regions of $\mathbf{K}$-space making important contributions to (25), both $(A^2-|B|^2)^{\frac{1}{2}}$ and $2SJK^2$ are small compared to $kT$. It is then permitted to expand the exponential in the integrand. Hence

$$M_T-M_T{}^{\text{Bloch}}=kT\frac{G_xG_yG_z}{V}\frac{2\beta}{(2\pi)^3}\int\left[\frac{A}{A^2-|B|^2}-\frac{1}{2SJK^2}\right]d\mathbf{K}. \tag{26}$$

From this point on, the integration is straightforward and yields:

$$M_T-M_T{}^{\text{Bloch}}=-\frac{G_xG_yG_z}{V}\frac{1}{S}\left(\frac{kT}{2SJ}\right)^{\frac{3}{2}}\frac{2\beta S}{(2\pi)^3}(2\pi)(2)(1.3)$$
$$\times\left\{\frac{3\pi}{8(1.3)}\left(\frac{2\beta H}{kT}\right)^{\frac{1}{2}}+\frac{\pi 2^{\frac{1}{2}}}{8(1.3)}\left(\frac{\beta}{kT}\right)^{\frac{1}{2}}\frac{H+4\pi M_0}{(4\pi M_0)^{\frac{1}{2}}}\sin^{-1}\left(\frac{4\pi M_0}{H+4\pi M_0}\right)^{\frac{1}{2}}\right\}. \tag{27}$$

As regards $M_\beta(H)$, it has not been found possible to evaluate the integral for it in a closed form for all values of $H$. However, in the two limits, $H\ll 4\pi M_0$ and $H\gg 4\pi M_0$, the integral has been evaluated. One obtains:

$$M_\beta(H)=\frac{G_xG_yG_z}{V}\beta\left(\frac{4\pi\beta M_0}{2SJ}\right)^2\left(\frac{2SJ}{2\beta H}\right)^{\frac{1}{2}}, \qquad \text{if } H\gg 4\pi M_0,$$

$$M_\beta(H)=\frac{G_xG_yG_z}{V}\beta\left(\frac{4\pi\beta M_0}{2SJ}\right)^2\left(\frac{2SJ}{4\pi\beta M_0}\right)^{\frac{1}{2}}, \quad \text{if } H\ll 4\pi M_0.$$

An estimate of the relative importance of the terms $M_\beta(H)$ and $M_T(T, H)$ is obtained by studying the change of these two quantities when $H$ varies from zero to, say, $4\pi\beta M_0$. Then

$$\Delta M_T\equiv M_T(T, 4\pi\beta M_0)-M_T(T, 0)\cong\frac{G_xG_yG_z}{V}\beta\left(\frac{kT}{2SJ}\right)^{\frac{3}{2}}\left(\frac{4\pi\beta M_0}{kT}\right)^{\frac{1}{2}},$$

$$\Delta M_\beta\equiv M_\beta(4\pi\beta M_0)-M_\beta(0)\cong\frac{G_xG_yG_z}{V}\beta\left(\frac{2SJ}{4\pi\beta M_0}\right)^{\frac{1}{2}}\left(\frac{4\pi\beta M_0}{2SJ}\right)^2$$

and

$$\Delta M_\beta/\Delta M_T=4\pi\beta M_0/kT. \tag{28}$$

This ratio is negligibly small ($5\times 10^{-3}$ for iron, $1.5\times 10^{-3}$ for nickel); hence, $M_\beta$ shall be omitted completely in what follows.

Thus, from (23), (24), and (27):

$$M(T, H)=M_0-\frac{G_xG_yG_z}{V}(2\beta S)\frac{1}{S}\left(\frac{kT}{2SJ}\right)^{\frac{3}{2}}\frac{(2\pi)(2)(1.3)}{(2\pi)^3}\left(1-\frac{3\pi}{8(1.3)}\left(\frac{2\beta H}{kT}\right)^{\frac{1}{2}}\right.$$
$$\left.-\frac{\pi}{8(1.3)}\left(\frac{8\pi\beta M_0}{kT}\right)^{\frac{1}{2}}\frac{H+4\pi M_0}{4\pi M_0}\sin^{-1}\left(\frac{4\pi M_0}{H+4\pi M_0}\right)^{\frac{1}{2}}\right). \tag{29}$$

The quantity most often found in the experiments is the "intrinsic susceptibility," $\partial M/\partial H$. One obtains from (29),

$$\frac{\partial M}{\partial H}=\frac{G_xG_yG_z}{V}(2\beta S)\frac{1}{S}\left(\frac{kT}{2SJ}\right)^{\frac{3}{2}}\frac{(2\pi)(2)(1.3)}{(2\pi)^3}\frac{1}{M_0}$$

$$\times\frac{1}{16(1.3)2^{\frac{1}{2}}}\left(\frac{4\pi\beta M_0}{kT}\right)^{\frac{1}{2}}\left(\left(\frac{4\pi M_0}{H}\right)^{\frac{1}{2}}+\sin^{-1}\left(\frac{4\pi M_0}{H+4\pi M_0}\right)^{\frac{1}{2}}\right). \quad (30)$$

Up to the present time, it has not proved feasible to calculate $J$ in terms of atomic constants; in practice its numerical value is obtained from an experimental measurement of $M_0-M(T,0)$. This latter quantity may be equated to $M_T{}^{\text{Bloch}}$ since the other contribution to $M_0-M(T,0)$, namely $M_T(T,0)-M_T{}^{\text{Bloch}}$ and $M_\beta$, is relatively small. Thus from (30) and (24), one finally obtains:

$$\frac{\partial M}{\partial H}=\left(\frac{M_0-M(T,0)}{M_0}\right)\left(\frac{1}{(16)(2^{\frac{1}{2}})(1.3)}\right)\left(\frac{4\pi\beta M_0}{kT}\right)^{\frac{1}{2}}\left(\left(\frac{4\pi M_0}{H}\right)^{\frac{1}{2}}+\sin^{-1}\left(\frac{4\pi M_0}{H+4\pi M_0}\right)^{\frac{1}{2}}\right). \quad (31)$$

The application of this result to the experiments is considered in Section VI.

It is instructive to compare the above result for $\partial M/\partial H$ with what would be obtained if the magnetic interactions were *not* treated quantum-mechanically, but classically by means of the Lorentz local field. The result of such a procedure can be obtained directly from (30) by

(1) Passing to the limit $M_0\rightarrow 0$.

(2) Replacing $H$ by $H+\gamma M$ where $\gamma$ is a numerical factor $\cong 4\pi/3$.

The constant $J$ can then be eliminated as above, and one obtains

$$\left(\frac{\partial M}{\partial H}\right)_{\text{Lorentz}}=\left(\frac{M_0-M(T,0)}{M_0}\right)\left(\frac{1}{(8)(1.3)(2^{\frac{1}{2}})}\right)\left(\frac{4\pi\beta M_0}{kT}\right)^{\frac{1}{2}}\left(\frac{4\pi M_0}{H+\gamma M_0}\right)^{\frac{1}{2}}. \quad (32)$$

The important differences between $(\partial M/\partial H)_{\text{Lorentz}}$ and $\partial M/\partial H$ of (31) occur for $H\ll 4\pi M_0$. In this case $(\partial M/\partial H)_{\text{Lorentz}}$ predicts a field-independent susceptibility, while (31) has an inverse square-root dependence on $H$. On the other hand for $H\gg 4\pi M_0$, the two expressions become identical; both then vary with $H$ as $H^{-\frac{1}{2}}$. The inverse square-root dependence of the susceptibility on the magnetic field for $H\gg 4\pi M_0$ is thus seen to be a general feature of the exchange interaction model subject to approximations I, II, III, quite independent of the details of the treatment of the magnetic interaction.

It should finally be noted from (30) that the susceptibility is proportional to the absolute temperature.

## V. Effects of Spin-Orbit Interactions

As was stated at the end of Section II, all spin-orbit effects have thus far been neglected in the formulation of the fundamental Hamiltonian (1). If one desires to treat these effects within the framework of the present development, i.e., supplementing the Hamiltonian (1) by additional terms, the first question which arises is whether spin-orbit effects can be described by an equivalent interaction energy in the space of the atomic spin variables. This question has been considered by Van Vleck in connection with the problem of ferromagnetic anisotropy.[11] In Van Vleck's work the spin-orbit effects are represented by equivalent "dipole-dipole" and "quadrupole-quadrupole" interactions of the type:

$$\tfrac{1}{2}\sum_{l,m}C_{lm}(R_{lm})[\mathbf{S}_l\cdot\mathbf{S}_m-3\overset{-2}{R_{lm}}\mathbf{S}_l\cdot\mathbf{R}_{lm}\mathbf{S}_m\cdot\mathbf{R}_{lm}], \tag{33}$$

$$\tfrac{1}{2}\sum_{l,m}\gamma_{lm}(R_{lm})\overset{-4}{R_{lm}}(\mathbf{S}_l\cdot R_{lm})^2(\mathbf{S}_m\cdot\mathbf{R}_{lm})^2. \tag{34}$$

These two terms are discussed in order.

(1) Conveniently, the equivalent "dipole-dipole" interaction (42) is just the same as the magnetic interaction provided that $4\beta^2/R^3{}_{lm}$ is replaced by $C_{lm}(R_{lm})$. The effect of this dipole-dipole interaction then merely consists of additions $\Delta A_\lambda$, $\Delta B_\lambda$ to the coefficients $A_\lambda$ which appear in the Hamiltonian (19), and therefore in the integral (23) for $M(T, H)$. One has: (cf. the first two sums in Eq. (12) for $A_\lambda$ and Eq. (13) for $B_\lambda$)

$$\Delta A_\lambda=\sum_h\{(-3S/2)C(R_h)(1-3z^2{}_h/R^2{}_h)+(S/2)C(R_h)(1-3z^2{}_h/R^2{}_h)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))\}, \tag{35}$$

$$\Delta B_\lambda=\sum_h\{(-3S/2)C(R_h)((x^2{}_h-y^2{}_h-2ix_hy_h)/R^2{}_h)\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h)\}. \tag{36}$$

It is also reasonable to suppose that $C(R_h)$ is of short range character;[17] in this case the sums in (35) and (36) have appreciable contributions only from the nearest neighbors of a given atom. Taking account of the crystalline cubical symmetry of the specimen, one obtains:

$$\Delta A_\lambda=\sum_h(S/2)C(1-3z^2{}_h/R^2{}_h)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h)),$$

$$\Delta B_\lambda=\sum_h(3S/2)C((x_h{}^2-y_h{}^2-2ix_hy_h)/R^2{}_h)(\mathbf{K}_\lambda\cdot\mathbf{R}_h)^2+\text{terms in }\mathrm{K}^4{}_\lambda,\ \mathrm{K}^6{}_\lambda,\ \cdots$$

with $C\equiv C(R_h)$ for nearest neighbors,[17a] $\cong 10$ wave-numbers $\cong J/100$.

Equations (35), (36) and the above numerical estimate for $C$ insure that the equivalent "dipole-dipole" interactions have no effect whatever on the magnetization. As far as the $A_\lambda+\Delta A_\lambda$ coefficient is concerned, the $\Delta A_\lambda$ term: $\sum_h(S/2)C(1-3Z^2{}_h/R^2{}_h)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))$ will be completely swamped by the exchange term in $A_\lambda$: $\sum_h 2SJ(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))$. On the other hand, the $B_\lambda+\Delta B_\lambda$ coefficient is altogether important only when it is $\cong A_\lambda+\Delta A_\lambda$, i.e., only when $4\pi\beta M_0\cong\sum_h 2SJ(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))\cong 2SJ\mathrm{K}^2{}_\lambda$. Under these circumstances, $\Delta B_\lambda\cong SC\mathrm{K}^2{}_\lambda\cong SC4\pi\beta M_0/2SJ\ll 4\pi\beta M_0$, and thus, $\Delta B_\lambda\ll B_\lambda$, for the important $B_\lambda$.

(2) Concerning the quadrupole-quadrupole interaction, an examination of its diagonal matrix element in the representation where the Hamiltonian of Eq. (19) is diagonal indicates that its effect, to the first order in $\gamma$, is given by the addition to $A_\lambda$ of a constant term $\cong\gamma$. This term may be conveniently described by a fictitious "magnetic field": $\mathrm{H}_a\equiv\gamma/2\beta\cong$anistropy constant$/M_0\cong 200$ gauss (Fe), 100 gauss (Ni).[17b] It can then be coupled together with the original $2\beta H$ term in $A_\lambda$ in the form $2\beta(H+H_a)$ and carried through to the end of the evaluation of $M(T, H)$, without further ado. Thus, in the expressions for $M$ and $\partial M/\partial H$ in Eqs. (30), (31), the term in $\gamma$ has but little effect if $H$ is, say, greater than 1000 guass.

(3) Finally, it is possible that some portion of the spin-orbit effects is incapable of description by interactions of the type (33), (34). Until explicit expressions are presented, one cannot, of course, estimate the resultant corrections. It is to be hoped, however, that these corrections will still be described by means of a fictitious field, $H_a$, whose order of magnitude will be the same as in the above numerical estimates.[17c]

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[17] Van Vleck, reference 11, p. 1193.

[17a] This numerical estimate for $C$ is due to Van Vleck, reference 11, p. 1184.

[17b] For experimental values of the anisotropy constant see Becker and Döring reference 3, p. 123, Table 12.

[17c] It may be remarked in passing that the methods of the present paper offer an approach to the problem of ferromagnetic anisotropy at low temperatures. For this purpose the eigenvalues $\mathrm{E}_{\mathrm{N}\lambda}$ of Eq. (20), with $B_\lambda$ replaced by $B_\lambda+\Delta B_\lambda$ must be determined as functions of the direction of $\mathbf{M}_0$ with respect to the crystallographic axes. This calculation and its relation to other treatments of ferromagnetic anisotropy [Van Vleck, reference 11; Van Peype, Physica **5**, 465 (1938)] is now being considered.

## VI. Comparison with Experiment

### (1) Numerical estimates of theoretical values for the susceptibility

Equation (31) for the susceptibility $\partial M/\partial H$, when applied to iron and nickel at $T=287°\mathrm{K}$ and $H=4000$ gauss, gives:

$$(\partial M/\partial H)_{\mathrm{Fe}}=1.7\times 10^{-4}, \tag{37}$$

$$(\partial M/\partial H)_{\mathrm{Ni}}=1.2\times 10^{-4}, \tag{38}$$

taking[18]

$$(M_0-M(T,0))/M_0]_{\mathrm{Fe}}=0.018, \qquad (4\pi M_0)_{\mathrm{Fe}}=21{,}800\,;$$

$$(M_0-M(T,0))/M_0]_{\mathrm{Ni}}=0.040, \qquad (4\pi M_0)_{\mathrm{Ni}}=6{,}400.$$

There exists no experimental data for cobalt for comparison with theory.

These numerical results for $\partial M/\partial H$, are to be compared with those of Akulov, which are based on the phenomenological Weiss-Heisenberg theory. In this theory,[19] for atoms with $S=\frac{1}{2}$,

$$M(T,H)=M_0\tanh\left[(\beta H+\beta qM)/kT\right],$$

$q=\frac{1}{2}zJ/\beta M_0$; whence, for low $T$, Akulov's results may be put in the form:

$$\left[\frac{\partial M}{\partial H}\right]_{\mathrm{Akulov}}=\frac{[M_0-M(T,0)]}{M_0}\frac{1}{2\pi}\frac{4\pi\beta M_0}{kT}. \tag{39}$$

The numerical values for iron and nickel given by (39) are found from the experimentally observed values of $[M_0-M(T,0)]/M_0$. One has:

$$\partial M/\partial H]_{\mathrm{Fe}}^{\mathrm{Akulov}}=1.5\times 10^{-5}, \tag{40}$$

$$\partial M/\partial H]_{\mathrm{Ni}}^{\mathrm{Akulov}}=1.2\times 10^{-5}. \tag{41}$$

Thus Akulov's susceptibility is numerically smaller than the one derived in this paper by a factor of about ten;[20] this factor is due essentially to the appearance in Eq. (39) of the quantity $4\pi\beta M_0/kT$, which is $\ll 1$, whereas in Eq. (31), there is instead the quantity $(4\pi\beta M_0/kT)^{\frac{1}{2}}$.

Further, Akulov's susceptibility is independent of $H$ and is proportional to $e^{-zJ/kT}/T$, while the susceptibility of Eq. (31), as has already been noted at the end of Section IV, is proportional to $TH^{-\frac{1}{2}}$, for a large range of fields.

### (2) Discussion of the various experiments

Information concerning the variation of the intrinsic magnetization with $H$, has been obtained from experiments[21] on polycrystalline samples of nickel and iron. In these experiments it has been customary to describe the variation of the observed magnetization with $H$ by a formula of the type:

$$(\partial M/\partial H)_{\mathrm{obs}}=A/H^2+B/H^3+C, \tag{42}$$

where $A$, $B$, $C$ are supposed to be constants.

The constants $A$ and $B$ depend upon the metallurgical history of the polycrystalline specimen; $A$ is conditioned by the plastic deformation of the material,[22] whereas $B$ is determined by the crystalline properties and elastic state.[22] The physical significance of the constant $A$ is at present a matter of speculation; the order of magnitude and dependence on temperature and elastic condition, of the constant $B$, as well as the field variation of the term $B/H^3$, shows that $B$ originates from the ferromagnetic anisotropy of the component crystal grains of the specimen.[23] The latter observation is true whether the anisotropy is a consequence of the natural crystalline properties, or is induced by an external stress. Finally, the constant $C$ has been

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[18] The values of $M_0$ for Fe and Ni are obtained from Becker and Döring, p. 27, Table 5. At $T=287°\mathrm{K}$, iron still obeys the $T^{\frac{3}{2}}$ law while nickel already shows some deviations. Under these circumstances one must take for $[(M_0-M(T,0))/M_0]_{\mathrm{Ni}}$ in Eq. (31), not the experimentally observed value, 0.054, but the value obtained by extrapolation of the $T^{\frac{3}{2}}$ law, the constant $2SJ$ in it being determined from low temperatures. The resulting value for $[(M_0-M(T,0))/M_0]_{\mathrm{Ni}}$ is 0.040. See the experimental work of Weiss and Forrer, and Fallot as quoted, e.g. in R. H. Fowler *Statistical Mechanics* (Cambridge University Press, 1936), p. 500.

[19] Cf. Reference 4. $q$ is the internal field constant and $z$ the number of nearest neighbors.

[20] Equation (39) and numerical values (40), (41), represent the maximum estimates which may be obtained for $\partial M/\partial H$ from the Weiss-Heisenberg phenomenological theory. Other estimates obtained by: (1) Use of theoretical value for $(M_0-M(T,0))/M_0$, i.e., $2e^{-zJ/kT}$. (2) Use of Langevin function appropriate to $S>\frac{1}{2}$, e.g. to $S=\infty$ — classical Weiss theory, give consistently smaller results.

[21] P. Weiss and R. Forrer, Ann. de physique **10**, 153 (1926); **12**, 279 (1929). E. Czerlinsky, Ann. d. Physik **13**, 89 (1932). A. R. Kaufmann, Phys. Rev. **55**, 1142, 1939 and private communication. H. Polley, Ann. d. Physik **36**, 625 (1939).

[22] A. R. Kaufmann, Phys. Rev. **51**, 1089 (1939).

[23] The theory of the term $B/H^3$ and of the constant $B$ has been given by Akulov, reference 3; the experimental verification by Kaufmann, Czerlinsky, and Polley, reference 21. See also, Becker and Döring, reference 3, pp. 167–176.

observed to be independent of the metallurgical treatment.[22]

The independence of $C$ from metallurgical treatment immediately suggests that it arises from the variation of the intrinsic domain magnetization with field. $C$ is thus to be compared with expression (31) for $\partial M/\partial H$ derived in this paper, and with the expression (39) for $\partial M/\partial H$ derived by Akulov.

(*a*) *Field dependence of* $C=\partial M/\partial H]_{\text{intrinsic}}$.—Unfortunately, all the experiments[21] except those of Weiss and Forrer have been carried out in such a range of fields that, for most of the range, the $A/H^2$ and $B/H^3$ terms both predominate over $C$. A typical example is the work of Polley,[21] where the range of fields in which $C$ is $\geqq A/H^2+B/H^3$ is from 3000–4000 gauss (cf. p. 641 of Polley's paper: Fig. 10). Under these conditions a weak dependence of $C$ on $H$ such as $H^{-\frac{1}{2}}$ could never be observed. All that can be obtained from these experiments, therefore, is the average numerical value of $C$ over a field range of about 2000–5000 gauss.

The only experiments for which the determination of the field dependence of $C$ is possible are those of Weiss and Forrer.[21] In these experiments the magnetization as a function of $H$ was determined in a range of fields up to 20,000 gauss. From 5000 to 20,000 gauss $C$ is the only term of importance; it was found to be independent of $H$. However, the slope of Weiss' $M$ *vs.* $H$ curves, i.e., $C$, is so small in absolute magnitude that it is difficult to decide whether this slope is actually a constant independent of $H$. In this connection, it should also be pointed out that Weiss' results have been criticized by other authors—particularly Czerlinsky;[21] *viz.*, Weiss measured directly $M$ as a function of $H$, with the result that small errors in the measurement of $M$ were capable of seriously affecting the determination of $\Delta M/\Delta H$; on the other hand, all the other experimenters measured $\Delta M/\Delta H$ directly. The relative inaccuracy of Weiss' procedure is indicated by the scattering of his observed values of $C$, i.e., from $0.72\times 10^{-4}$ to $1.2\times 10^{-4}$.

(*b*) *Magnitude of* $C$.—The most consistent values of $C$ for Ni have been obtained by Polley.[21] For $T=287°\text{K}$ and $H=4000$ gauss, Polley finds $C$ to be $1.3\times 10^{-4}$, which is in excellent agreement with Eq. (38) of this paper, but which is ten times larger than the value of $C$ derived by Akulov [cf. Eq. (41)]. Other values of $C$ have been obtained by Kaufmann;[21] these are $C=1.0\times 10^{-4}$ and $C=0.7\times 10^{-4}$.

For iron there are available the results of Kaufmann[21] and Czerlinsky;[21] Kaufmann obtained $C=3.8\times 10^{-4}$ and $4.8\times 10^{-4}$, while Czerlinsky found $C=4.4\times 10^{-4}$. The average of these results[24] is about two and a half times the theoretical value found in this paper [cf. Eq. (37)] and more than twenty-five times Akulov's theoretical value [cf. Eq. (40)].

(*c*) *Temperature dependence of* $C$.—The expression (30) for $\partial M/\partial H$ predicts a linear variation of $C$ with temperature. This is experimentally verified by Polley[21] in the temperature range 250°K–320°K.[25]

### (3) Further possible experiments

A great complication in the experiments so far performed arises from the presence of the terms $A/H^2$ and $B/H^3$, which are due to the polycrystalline nature of the specimen. It would therefore be desirable to obtain experimental data with single crystals and with $H$ in the direction of the various axes of symmetry in the crystal. These directions may be either the directions of "easy" or of "difficult" magnetization. In the first case, technical saturation is achieved for $H\gtrapprox 50$ gauss;[26] in the second case, technical saturation is achieved for $H\gtrapprox 3H_a\cong 600$ gauss (Fe), 300 gauss (Ni).[27] Any further observed variation of $M$ with $H$, beyond its technical saturation value, is then to be ascribed to changes in the intrinsic magnetization. Thus, using single crystals, one may obtain, directly from the $\partial M/\partial H$ *vs.* $H$ curves, the various features of theoretical interest.

(*a*) *Functional dependence of* $\partial M/\partial H$ *on* $H$, *with* $H\gg 4\pi M_0$: Test of the $H^{-\frac{1}{2}}$ dependence.[28]

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[24] Dr. Kaufmann, in a private communication, has stated that there exists a large experimental error (*ca.* 100 percent) in his determination of $C$.

[25] At higher temperatures, deviations from the linear variation of $\partial M/\partial H$ with $T$ are to be expected, and have been found by Polley, reference 21. These deviations correspond to departures from the $T^{\frac{3}{2}}$ law in the same temperature range.

[26] See Becker and Döring, reference 3, p. 102, Fig. 61; p. 118, Fig. 71.

[27] Becker and Döring, reference 3, p. 102, Fig. 61; p. 118, Fig. 71; p. 124, Fig. 77.

[28] To determine experimentally the variation of $\partial M/\partial H$ with $H$, for $H\gg 4\pi M_0$, one may also use polycrystalline specimens, since for such high fields the terms $B/H^3$ and $A/H^2$ are negligible.

(*b*) *Variation of $\partial M/\partial H$ with $H$* for $3H_a < H \leqq 4\pi M_0$: Test of effect of magnetic interaction in Eq. (31).

(*c*) *Effect of spin-orbit interactions:* With $H$ parallel to the direction of "easy" magnetization, an investigation of the variation of $\partial M/\partial H$ with $H$, for $50 < H < 3H_a$, may be undertaken. There are then three possibilities:

(1) The spin-orbit effects are entirely described by the equivalent "dipole-dipole" interaction (33). In this case, the $\partial M/\partial H$ *vs.* $H$ curves in this field range should be described by Eq. (31).

(2) An appreciable part of the spin-orbit effects is due to the equivalent "quadrupole-quadrupole" interaction (34). The $\partial M/\partial H$ *vs.* $H$ curves in this field range should now be given by Eq. (31), with $H$ replaced by $H+H_a$.

(3) An appreciable part of the spin-orbit effects cannot be described by equivalent interactions of the type (33), (34). The $\partial M/\partial H$ *vs.* $H$ curves in this field range may then show a more radical departure from Eq. (31).

### (4) Inadequacy of other possible mechanisms to account for the observed variation of the intrinsic magnetization with field

These mechanisms are:

(*a*) Ion-core diamagnetism of the atoms: resulting volume susceptibility, $\partial M/\partial H$ is $\lesssim 10^{-6}$, and is negligible.

(*b*) Spin paramagnetism and orbital diamagnetism of the conduction electrons: resulting volume susceptibility $\partial M/\partial H$, is $\lesssim 10^{-6}$, and is negligible.[29]

(*c*) Paramagnetism arising from the orbital motions of the electrons in the incomplete $d$ shells. (The exchange coupling of the spins of these electrons gives rise to the ferromagnetism.) Precise estimates of the order of magnitude of this paramagnetism are difficult since little is known about the orbital states. Assuming that the orbital moment, $\mathbf{L}$, of the incomplete $d$ shell is what it would be in the vapor state, and that the various orbital magnetic moments do not at all influence each other, one obtains, with the usual Curie law:

$$\partial M/\partial H = (N/V)(\beta^2 L(L+1)/3kT),$$

the following paramagnetic volume susceptibilities: $3\times 10^{-4}$ for iron; $7\times 10^{-4}$ for nickel. The gyromagnetic experiments, on the other hand, indicate that:

Either the orbital angular momentum of each incomplete $d$ shell is destroyed or quenched by the crystalline electric field,[30a] in which case the corresponding susceptibility vanishes in first order;

Or, the orbital angular momentum vectors of the different atoms are "antiferromagnetically coupled." [30b] Then an estimate of the susceptibility may be obtained by replacing $kT$ in the Curie law above by $kT+kT_c$, where $kT_c$, the interaction energy of the antiferromagnetic coupling, is $\lesssim 1\frac{1}{4}$ ev.[31] The resulting paramagnetic volume susceptibility is thus cut down by a factor of fifty, and becomes negligibly small.

In conclusion, it should also be noted that the smallness of any paramagnetic susceptibility, described by the Curie-Weiss law, is directly shown by the experimentally observed temperature variation of the intrinsic magnetization; *viz.*, $C = \partial M/\partial H]_{\text{intrinsic}} = \text{const. } T$ (see Section VI).

The authors wish to thank Dr. A. R. Kaufmann of the Massachusetts Institute of Technology for helpful discussions of the experiments, and for communication of experimental data before publication.

## Appendix I

The various approximations made at the beginning of the treatment are of the following type:

I. Neglect of $a^*_l(1-a^*_l a_l/2S)^{\frac{1}{2}} - a^*_l$ in comparison with $a^*_l$.

II. Neglect of $a^*_l a_l a^*_m a_m/2S \equiv \mathfrak{n}_l \mathfrak{n}_m/2S$ in comparison with $a^*_l a_l \equiv \mathfrak{n}_l$.

III. Neglect of $a^*_m a_m a^*_l \equiv \mathfrak{n}_m a^*_l$ in comparison with $a^*_l a^*_m$.

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[29] In making this estimate, it is assumed that the conduction electrons are perfectly free. This assumption is reasonable since the conduction electrons are valence electrons arising from the 4*s* atomic shells. The strong paramagnetism, with volume susceptibility $\lesssim 5\times 10^{-5}$, arising from electrons in the incomplete inner shells of some of the nonferromagnetic transition elements, e.g. Pd, plays no role in ferromagnetic elements, since the incomplete inner shells in this case, are responsible for the ferromagnetism itself.

[30a] Van Vleck, reference 4, Ch. 11.

[30b] Van Vleck, reference 11, p. 1182.

[31] Van Vleck, reference 11, p. 1184.

The neglected terms are important whenever two or more spin deviations are in close proximity, since all the neglected terms in I, II, III are multiplied by short range functions of $R_{lm}$ i.e. $J_{lm}(R_{lm})$ or $R^{-3}{}_{lm}$. When spin deviations are to be found on atoms close to each other, the following situations may arise:

(A) The system makes transitions from states with $n_l \geqq 1$ to states with $n_l+1$. Cf. I, i.e.,

$$[a^*{}_l(1-a^*{}_l a_l/2S)^{\frac{1}{2}}-a^*{}_l]\Psi_{n_l}$$
$$=(n_l+1)^{\frac{1}{2}}((1-n_l/2S)^{\frac{1}{2}}-1)\Psi_{n_l+1}=0,$$

unless $n_l \geqq 1$.

(B) The system occupies states with $n_l>0$, $n_m>0$. Cf. II, i.e.,

$$\frac{\mathfrak{n}_l\mathfrak{n}_m}{2S}\Psi_{n_l n_m}=\frac{n_l n_m}{2S}\Psi_{n_l n_m}.$$

(C) The system makes transitions from states with $n_m>0$, $n_l$ to states with $n_m$, $n_l+1$. Cf. III, i.e.,

$$\mathfrak{n}_m a^*{}_l\Psi_{n_l n_m}=n_m(n_l+1)^{\frac{1}{2}}\Psi_{n_l+1,\, n_m}.$$

It is thus seen that I, II, III, imply that any atom having a spin deviation can always be regarded as surrounded by atoms with no spin deviations, i.e., the spin deviations are regarded as isolated independent units with no interactions.

Now, spin deviations may come into close proximity and give rise to (A), (B), and (C), as a result of two mechanisms, The first of these is temperature agitation, the second, the action of terms in the magnetic interaction of the form $R^{-3}{}_{lm}a^*{}_l a_m$ in creating two spin deviations close to each other.

In view of the lack of knowledge of the wave functions of the exact Hamiltonian (6) or (1), the approximate Hamiltonian (7) (which is the exact Hamiltonian (6) or (1), subject to approximation I, II, III) will be used to estimate the relative importance of the cases of close proximity of spin deviations.

(1) *Temperature agitation.*—The structure of the Hamiltonian (7), (without the part of the magnetic interaction proportional to $a^*{}_l a^*{}_m$ and $a_l a_m$) or better the Hamiltonian (11), (without the terms in $B_\lambda$ and $B^*{}_\lambda$) obtained from (7) by the Fourier transformation (8), indicates that the position of each spin deviation is spatially uncorrellated with the position of any of the other spin deviations. Thus cases of close proximity of spin deviations due to temperature agitation are relatively infrequent in the condition of quasi-saturation.

To verify this lack of spatial correlation, one observes that

$$\langle\langle\mathfrak{n}_l\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}\equiv\langle\langle a^*{}_l a_l a^*{}_m a_m\rangle\rangle_{\mathrm{Av}}$$
$$=N^{-2}\sum_{\mu,\,\nu,\,\lambda,\,\rho}\exp\,\{i[(\mathbf{K}_\nu-\mathbf{K}_\mu)\cdot\mathbf{R}_l+(\mathbf{K}_\rho-\mathbf{K}_\lambda)\cdot\mathbf{R}_m]\}\langle\langle a^*{}_\mu a_\nu a^*{}_\lambda a_\rho\rangle\rangle_{\mathrm{Av}}$$
$$=N^{-2}\textstyle\sum_{\mu,\,\lambda}\langle\langle\mathfrak{n}_\mu\mathfrak{n}_\lambda\rangle\rangle_{\mathrm{Av}}+N^{-2}\sum_{\mu,\,\lambda}\exp\,\{i(\mathbf{K}_\mu-\mathbf{K}_\lambda)\cdot\mathbf{R}_h\}\langle\langle\mathfrak{n}_\mu(\mathfrak{n}_\lambda+1)\rangle\rangle_{\mathrm{Av}}(1-\delta_{\mu\lambda}).$$

The form of the partition function $Z$ in Eq. (22) (with $B_\lambda=0$, and hence $\mathrm{N}_\lambda=n_\lambda$), which is involved in averaging the $\mathfrak{n}_\lambda$ gives:

$$\langle\langle\mathfrak{n}_\mu\mathfrak{n}_\lambda\rangle\rangle_{\mathrm{Av}}=\langle\langle\mathfrak{n}_\mu\rangle\rangle_{\mathrm{Av}}\langle\langle\mathfrak{n}_\lambda\rangle\rangle_{\mathrm{Av}}(1-\delta_{\mu\lambda})+(\langle\langle\mathfrak{n}_\mu\rangle\rangle_{\mathrm{Av}}+2\langle\langle\mathfrak{n}_\mu\rangle\rangle_{\mathrm{Av}}{}^2)\delta_{\mu\lambda}.$$

Hence,

$$\langle\langle\mathfrak{n}_l\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}=N^{-2}\textstyle\sum_{\mu,\,\lambda}\langle\langle\mathfrak{n}_\mu\rangle\rangle_{\mathrm{Av}}\langle\langle\mathfrak{n}_\lambda\rangle\rangle_{\mathrm{Av}}+N^{-2}\sum_{\mu,\,\lambda}\exp\,\{i(\mathbf{K}_\mu-\mathbf{K}_\lambda)\cdot\mathbf{R}_h\}\langle\langle\mathfrak{n}_\mu\rangle\rangle_{\mathrm{Av}}\langle\langle\mathfrak{n}_\lambda\rangle\rangle_{\mathrm{Av}}.$$

Also

$$S[M_0-M(T,\,0)]/M_0=\langle\langle n_l\rangle\rangle_{\mathrm{Av}}=N^{-1}\textstyle\sum_m\langle\langle\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}=N^{-1}\sum_\mu\langle\langle\mathfrak{n}_\mu\rangle\rangle_{\mathrm{Av}}.$$

Thus $\langle\langle\mathfrak{n}_l\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}$ and $\langle\langle\mathfrak{n}_l\rangle\rangle_{\mathrm{Av}}\langle\langle\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}$ are both of the same order of magnitude, and hence, there are no spatial correlations among the spin deviations. It follows that the probability for a spin deviation to be found on atom $l$ when another spin deviation is known to be on atom $m$, is independent of the distance between atoms $l$ and $m$, and is $\cong\langle\langle\mathfrak{n}_l\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}/\langle\langle\mathfrak{n}_m\rangle\rangle_{\mathrm{Av}}\cong\langle\langle\mathfrak{n}_l\rangle\rangle_{\mathrm{Av}}\cong S[M_0-M(T,\,0)]/M_0\ll 1$ in the condition of quasi-saturation.

(2) *Magnetic interactions proportional to* $a^*{}_l a^*{}_m$. —The effect of these magnetic interactions is contained in the terms, $\sum_\lambda \frac{1}{2} B^*{}_\lambda a^*{}_\lambda a^*{}_{-\lambda}$, of the Hamiltonian (11).

Now the $B_\lambda$ which make the greatest contributions to $M(T,H)$ have $|\mathbf{K}_\lambda| \lesssim (4\pi\beta M_0/2SJ)^{\frac{1}{2}}$ [cf. text after Eq. (25)]. Also the main contributions to $B_\lambda$, (cf. (13)), come from summands for which $|\mathbf{R}_h|$, the relative separations of the spin deviations, is $\lesssim 1/\mathrm{K}_\lambda$. Thus the effective spatial separation of the spin deviations created by the magnetic interaction is $\cong (2SJ/4\pi\beta M_0)^{\frac{1}{2}}$ lattice spacings $\cong 30$ lattice spacings. However the average spatial separation of the spin deviations created by the temperature agitation is $\cong (M_0/[M_0 - M(T, 0)])^{\frac{1}{3}}$ lattice spacings $\cong 4$ lattice spacings. Thus from the point of view of causing cases of close proximity of spin deviations, the magnetic interaction is much less important than the temperature agitation.[32]

It should be emphasized that the above estimates as to the effective separation of the spin deviations created both by temperature agitation and by the magnetic interaction is given on the basis of the approximate Hamiltonian (7). The final justification of the validity of these estimates awaits a better knowledge of the wave functions of the exact Hamiltonian (6) or (1) than exists at present.

## APPENDIX II

The sums for $A_\lambda$ and $B_\lambda$ in (12) and (13) will now be evaluated. The contribution to the first sum in $A_\lambda$, arising from near neighbors of a given atom vanishes because of the cubical symmetry of the specimen. A small sphere may now be drawn enclosing all these near neighbors, and the contributions to the sum for atoms lying outside of this sphere replaced by contributions to the corresponding integral, in accordance with the prescription:

$$a^{-3} \sum_h 2\beta S f(\mathbf{R}_h) \to M_0 \int f(\mathbf{R}) d\mathbf{R}. \tag{43}$$

Thus,

$$\sum_h (-2\beta S/a^3 R^3{}_h)(1 - 3z^2{}_h/R^2{}_h) \to (-M_0) \int [(1 - 3z^2/R^2)/R^3] d\mathbf{R}, \tag{44}$$

where the integral is extended over all of the space outside the small sphere and inside the outer boundary of the specimen. Now by the divergence theorem,

$$(-M_0) \int [(1-3z^2/R^2)/R^3] d\mathbf{R} = (-\mathbf{M}_0) \cdot \int (\mathbf{n}z/R^3) d\Sigma + (-\mathbf{M}_0) \cdot \int (\mathbf{n}z/R^3) d\Sigma, \tag{45}$$

where, on the right-hand side, the first integral is extended over the surface of the small sphere, and the second integral over the outer boundary of the specimen, $\mathbf{n}$ being the outer normal to the surfaces. The integral over the surface of the small sphere gives $4\pi M_0/3$ and is thus just the Lorentz local field. The second integral depends on the shape of the outer boundary of the specimen and is just the demagnetizing field. One takes the specimen to be an ellipsoid whose major axis is the $z$ axis, and which is so elongated that the demagnetizing field may be neglected. Then,

$$3\beta \sum_h (-2\beta S/a^3 R^3{}_h)(1 - 3z^2{}_h/R^2{}_h) = 4\pi\beta M_0. \tag{46}$$

The second sum in $A_\lambda$ may be replaced by an integral at once, since this sum has no singularity at $\mathbf{R}_h = 0$. Thus:

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[32] The portion of the equivalent dipole-dipole interaction of the form $C(R_{lm})a^*{}_l a^*{}_m$ (cf. Section V) creates spin deviations on atoms which are nearest neighbors, since $C$ is short range. Nevertheless, the *effective* spatial separation of the spin deviations created by the *sum* of the magnetic and equivalent dipole-dipole interactions, *viz.* by terms of the type, $\sum_\lambda \frac{1}{2}(B^*{}_\lambda + \Delta B^*{}_\lambda) a^*{}_\lambda a^*{}_{-\lambda}$, is not appreciably different from 30 lattice spacings, since for $|\mathbf{K}| \lesssim (4\pi\beta M_0/2SJ)^{\frac{1}{2}}$, the values of $|\mathbf{K}_\lambda|$ where $B_\lambda + \Delta B_\lambda$ as well as $B_\lambda$ makes its most important contribution to $M(T, H)$, one has $\Delta B_\lambda \cong SC\mathrm{K}_\lambda{}^2 \cong (4\pi\beta M_0)(C/J) \ll 4\pi\beta M_0$. Thus, the contribution of $\Delta B_\lambda$ to the term $B_\lambda + \Delta B_\lambda$ is so small that the *effective* spatial separation of the spin deviations remains essentially unaltered.

$$\beta \textstyle\sum_h (2\beta S/a^3R^3{}_h)(1-3z^2{}_h/R^2{}_h)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))$$

$$\rightarrow \beta M_0\int((1-3z^2/R^2)/R^3)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}))d\mathbf{R}=i\beta M_0\mathrm{K}^{(z)}{}_\lambda\int(z/R^3)(\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}))d\mathbf{R} \quad (47)$$

by an integration by parts.

The last integral is just equal to $4\pi i\mathrm{K}^{(z)}{}_\lambda/\mathrm{K}^2{}_\lambda$; (12), (46), (47) then yield:

$$A_\lambda=A_{-\lambda}=4\pi\beta M_0\sin^2\theta_\lambda+\textstyle\sum_h 2SJ_h(R_h)(1-\exp(i\mathbf{K}_\lambda\cdot\mathbf{R}_h))+2\beta H. \quad (48)$$

To evaluate $B_\lambda$ one proceeds similarly. Considering the first sum in $B_\lambda$, one draws a small sphere about $\mathbf{R}_h=0$, enclosing all the near neighbors. The contributions to the sum from atoms within the sphere is again zero because of cubical symmetry, while the contributions from atoms outside the small sphere are calculated by replacing the sum with the corresponding volume integral. Thus,

$$-3\beta\textstyle\sum_h(2\beta S/a^3R^3{}_h)(x^2{}_h-y^2{}_h-2ix_hy_h)/R^2{}_h\rightarrow -3\beta M_0\int[(x^2-y^2-2ixy)/R^5]d\mathbf{R}$$

$$=3\beta M_0\int[(\mathbf{a}_x-i\mathbf{a}_y)\cdot\mathbf{n}(x-iy)R^{-3}]d\Sigma+3\beta M_0\int[(\mathbf{a}_x-i\mathbf{a}_y)\cdot\mathbf{n}(x-iy)R^{-3}]d\Sigma, \quad (49)$$

where the first surface integral is extended over the boundary of the small sphere, the second over the outer boundary of the specimen, and $\mathbf{a}_x$, $\mathbf{a}_y$ are unit vectors in the $x$ and $y$ directions. Also the integral over the small sphere's surface vanishes by symmetry.

The second sum in $B_\lambda$, just like the second sum in $A_\lambda$ may be replaced at once by an integral, which when integrated by parts gives:

$$-3\beta M_0\int[(\mathbf{a}_x-i\mathbf{a}_y)\cdot\mathbf{n}(x-iy)R^{-3}]d\Sigma$$
$$+4\pi\beta M_0(\mathrm{K}^{(x)}{}_\lambda-i\mathrm{K}^{(y)}{}_\lambda)^2/\mathrm{K}^2{}_\lambda, \quad (50)$$

where the surface integral is extended over the outer boundary of the specimen. Thus from (13), (49), (50)

$$B_\lambda=B_{-\lambda}=4\pi\beta M_0(\mathrm{K}^{(x)}{}_\lambda-i\mathrm{K}^{(y)}{}_\lambda)^2/\mathrm{K}^2{}_\lambda$$
$$=4\pi\beta M_0\sin^2\theta_\lambda e^{-2i\phi_\lambda}. \quad (51)$$

It should finally be stated that the replacement of sums by integrals in $A_\lambda$, $B_\lambda$ is an approximation with an error $\cong(\mathrm{K}_\lambda)^2$. This introduces an error into $M(T,H)\cong 4\pi\beta M_0/2SJ\ll 1$, since the greatest contributions to $M(T,H)$ from the magnetic interactions, both in $A_\lambda$ and $B_\lambda$ come from regions of $\mathbf{K}_\lambda$-space, where $|\mathbf{K}_\lambda|\lesssim(4\pi\beta M_0/2SJ)^{\frac{1}{2}}\ll 1$. [Cf. text after Eq. (25).]

The difference between the sums and integrals becomes much more important for problems of other types: e.g., *can ferromagnetism arise from pure magnetic dipole-dipole interactions?*[33]

In this case $J=0$, and the use of (48), (51),

$$A_\lambda=4\pi\beta M_0\sin^2\theta_\lambda+2\beta H;$$
$$B_\lambda=4\pi M_0\sin^2\theta_\lambda e^{-2i\phi_\lambda},$$

leads to a negative answer to the proposed question. It is to be expected, however, that the exact expressions for $A_\lambda$ and $B_\lambda$ determined by evaluation of the original sums (12), (13), will not be given by (48), (51), hence the existence of ferromagnetism arising from magnetic dipole-dipole interactions cannot be determined from the knowledge of $A_\lambda$ and $B_\lambda$ available at this time. The evaluation of the sums for $A_\lambda$ and $B_\lambda$ is being considered at present.

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[33] This problem has been discussed by J. H. Van Vleck, J. Chem. Phys. **5**, 320 (1937), Section 5; J. A. Sauer, Phys. Rev. **57**, 142 (1940).