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Theory of Critical Scattering

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1. INTRODUCTION

A SUBSTANCE which undergoes an order-disorder transition of second degree shows, near the critical temperature, a large increase in its cross section for the scattering of waves. This is due to the large fluctuations about the average which occur in this temperature region. Probably the best known example is the phenomenon of critical opalescence caused by the scattering of visible light in a liquid or dense gas near critical conditions. Considerable interest has recently been aroused by the observation of a large increase in the magnetic scattering of neutrons from ferromagnets close to their Curie points which may be attributed to a similar effect. Most of the studies of this so-called critical scattering have been on iron where the effect has been observed in total cross section by Palevsky and Hughes[1] and by Squires,[2] around the forward direction by Shull, Wilkinson, and Gersch,[3,4] and near a Bragg reflection by Lowde.[5] Similar effects appear in magnetite,[6] and should be observable in antiferromagnets and other substances with more complicated magnetic order under appropriate conditions. They should also be found in alloys which show order-disorder transitions of second degree with both x-rays and neutrons, but have not so far been observed.

The theoretical discussion of critical scattering in the liquid-gas system was first given by Ornstein and Zernike[7] some fifty years ago, and their method has been generalized by Klein and Tisza.[8] Van Hove[9,10] pointed out the origin of the magnetic critical scattering and extended the theory to cover this case. The theories determine the cross section as a Fourier transform of the so-called pair correlation function of particles and spins. The general form and size of the effect was, however, only determined to order of magnitude by using thermodynamic arguments. A determination of the scattering in terms of a microscopic theory is given in this paper so that the detailed observations[1–5] now available may be interpreted in terms of standard theories of order in alloys and magnets. The method is an extension of the Bethe-Peierls[11] (B-P) and other closely related methods which give a good treatment of order-disorder problems near and above the critical temperature. The Ising model is used for alloys and a crude treatment of magnets, which are also examined using the Heisenberg model. There is a model for the gas-liquid transition which is equivalent to the Ising model,[12] namely, the lattice gas or hole theory but it is a much cruder approximation in this case and will not be considered in detail.

The method is an improvement and extension of the calculation by Zernike[13] of the propagation of correlation in alloys near T_c. The approximation is also reasonably satisfactory at high temperatures but fails at low temperatures where the correlations have been discussed by other methods.[13,10,14]

2. SCATTERING FORMULAS

Van Hove has shown that the concept of the pair correlation function which is so useful in discussing the properties of x-ray scattering,[15] can be extended to cover the cases of interest here. Because the energy of neutrons used in diffraction experiments of this kind (i.e., neutrons with wavelength of order 1 to 10 A) is small, the energy changes which take place on scattering have an important effect on the cross section. This can be taken into account by considering the time dependence of the correlation function. Also, because the spins are vectors, it is necessary to define pair correlation functions for their components in the principal directions. Thus Van Hove defines[10]

$$\Gamma^{\alpha\beta}(\mathbf{r},t)=N^{-1}\Big\langle\sum_{i,j}\int d\mathbf{r}'\mathbf{S}_i^{\alpha}(0)\delta(\mathbf{r}+\mathbf{r}_i(0)-\mathbf{r}')\times\mathbf{S}_j^{\beta}(t)S(\mathbf{r}'-\mathbf{r}_j(t))\Big\rangle \quad (2.1)$$

the sum being over all pairs i, j of N spins. The position and α component of one spin are measured at time t before the position and β component of the other. The differential cross section per atom per unit solid angle

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[2] G. L. Squires, Proc. Phys. Soc. (London) **A67**, 248 (1954).

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[7] L. S. Ornstein and F. Zernike, Proc. Amsterdam Acad. Sci. **17**, 793 (1914).

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[9] L. Van Hove, Phys. Rev. **93**, 202 (1954).

[10] L. Van Hove, Phys. Rev. **93**, 1374 (1954).

[11] H. A. Bethe, Proc. Roy. Soc. (London) **A150**, 552 (1935); R. Peierls, Proc. Cambridge Phil. Soc. **32**, 477 (1936).

[12] T. D. Lee and C. N. Yang, Phys. Rev. **87**, 410 (1952).

[13] F. Zernike, Physica **7**, 565 (1940).

[14] J. Ashkin and W. E. Lamb, Jr., Phys. Rev. **63**, 159 (1946).

[15] See, for example, N. S. Gingrich, Revs. Modern Phys. **15**, 90 (1943).

and per unit energy of the scattered neutron is related to the Fourier transform of Γ. For unpolarized neutrons this is

$$\frac{d^2\sigma}{d\Omega dE}=\left(\frac{2ge^2}{mc^2}\right)^2\frac{k}{hk_0}|F(\boldsymbol{\kappa})|^2\sum_{\alpha\beta}(\delta_{\alpha\beta}-e_\alpha e_\beta)$$
$$\times\int\Gamma^{\alpha\beta}(\mathbf{r},t)\exp(i(\boldsymbol{\kappa}\cdot\mathbf{r}-\omega t))d\mathbf{r}dt,\quad(2.2)$$

where $\mathbf{k}_0$, $\mathbf{k}$ are the incident and scattered neutron wave vectors and $\boldsymbol{\kappa}=\mathbf{k}-\mathbf{k}_0$ is the scattering vector. $\hbar\omega$ is the energy charge on scattering $\hbar^2(k_0^2-k^2)/2m_0$, $g(=1.91)$ is the neutron's magnetic moment, m the electron, and m_0 the neutron mass. $F(\boldsymbol{\kappa})$ is the form factor which is the Fourier transform of the spin density distribution in each atom, and falls off like $e^{-\kappa\rho}$ as κ increases (where ρ is the atomic radius). $\mathbf{e}$ is the unit vector in the direction of $\boldsymbol{\kappa}$. The factor in $\mathbf{e}$ arises from the anisotropic nature of the electron-neutron spin-spin interaction.[16]

In the magnetic models considered below each spin is always found on a particular atom with equilibrium positions on lattice points $\mathbf{R}$. The thermal motion of the atoms around these positions is, to a good approximation, independent of the time dependence of the spin components. Therefore (2.1) may be written

$$\Gamma^{\alpha\beta}(\mathbf{r},t)=\sum\nolimits_{\mathbf{R}}\gamma^{\alpha\beta}(\mathbf{R},t)G_{\mathbf{R}}(\mathbf{r},t),\quad(2.3)$$

where

$$\gamma^{\alpha\beta}(\mathbf{R},t)=\langle\mathbf{S}^\alpha(0,0)\mathbf{S}^\beta(\mathbf{R},t)\rangle.\quad(2.4)$$

In the ordered phase the average components of spin on each atom are not all zero so that it is convenient to write

$$\gamma^{\alpha\beta}(\mathbf{R},t)=\langle\mathbf{S}^\alpha\rangle\langle\mathbf{S}^\beta\rangle+\gamma'^{\alpha\beta}(\mathbf{R},t).\quad(2.5)$$

The first term representing the long range order gives elastic Bragg scattering, which in a crystal with all spins ordered in the z direction is

$$\frac{d\sigma}{d\Omega}=\frac{NV_0}{(2\pi)^3}\left(\frac{2ge^2}{mc^2}\right)^2$$
$$\times|F(\boldsymbol{\kappa})|^2(1-e_z^2)\langle\mathbf{S}^z\rangle^2e^{-2W}\sum_{\tau}\delta(\boldsymbol{\kappa}-\boldsymbol{\tau}),\quad(2.6)$$

where $\boldsymbol{\tau}$ is a reciprocal lattice vector, and the effect of the thermal motion on the atom position introduces the Debye-Waller factor e^{-2W}. Here, V_0 is the volume of a unit cell. In magnets with more complex order, the appropriate reciprocal lattice will enter and a structure factor will replace $\langle S^z\rangle^2$. The second term of (2.5) gives an additional diffuse scattering which is in general inelastic and is given by (1.2) with γ' replacing Γ and the Debye-Waller factor included.†

The calculation of critical magnetic scattering therefore reduces to a calculation of $\gamma'^{\alpha\beta}(\mathbf{R},t)$ at temperatures close to the critical point. This is a convenient formulation since γ' is an extension to all $\mathbf{R}$, t of the short range order parameters which are often discussed by order-disorder theories. It is convenient to consider first the instantaneous correlation function $\gamma^{\alpha\beta}(\mathbf{R},0)$ and we shall confine our attention to this static approximation. The time dependence, and hence, the inelasticity of the scattering will be considered in a further paper. The quantity $\gamma'^{\alpha\beta}(\mathbf{R},0)$ is interesting in itself and useful under certain conditions since, if energy transfers $\hbar\omega$ are small compared to the incident energy, one may write

$$\int\Gamma(\mathbf{r},t)\exp(i(\boldsymbol{\kappa}\cdot\mathbf{r}-\omega t)dt\simeq\delta(\omega)\Gamma(\mathbf{r},0)\exp(i\boldsymbol{\kappa}\cdot\mathbf{r}).\quad(2.7)$$

This static approximation is always valid for x-rays since they have such large energies. Close to the critical point the scattering becomes predominantly elastic[10] so that it is actually a useful approximation in the neutron case also.

3. ISING MODEL OF A FERROMAGNET $S=\frac{1}{2}$

As the simplest case which allows a clear discussion of the method, the Ising model of a ferromagnet with $S=\frac{1}{2}$ is first considered, although it is not a realistic model for any physical case. It may, however, be readily extended, as shown in later sections.

In the Ising model attention is confined to the z components of the spins, with the two possible values $\pm\frac{1}{2}$. The interaction potential between two spins is replaced by

$$-2JS^z(\mathbf{R})S^z(\mathbf{R}+\boldsymbol{\beta}),\quad(3.1)$$

where $\boldsymbol{\beta}$ defines the nearest neighbor positions of $\mathbf{R}$. Also attention is confined to those cubic lattices where two nearest neighbors of a given atom are not nearest neighbors of each other, namely, the simple cubic where the number of neighbors is $z=6$ and the body-centered cubic where $z=8$. In each case the side of the cubic cell is taken to be a. In the face-centered lattice ($z=12$), where this condition is not fulfilled, an extension of the usual Bethe-Peierls type of theory is required. The present method of discussing γ could also be extended to this case but is not treated in this paper.

The B-P method[17] treats the order-disorder problem

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[16] O. Halpern and M. H. Johnson, Phys. Rev. 55, 898 (1939).

† The time dependence of $G(r,t)$ also allows the neutron to exchange energy with the thermal vibrations. Through this, the first term in (2.5) gives a cross section for phonon scattering in addition to the usual phonon scattering from the neutron-nuclear interaction. This magnetovibrational scattering [R. J. Elliott and R. D. Lowde, Proc. Roy. Soc. (London) A230, 46 (1955)] is sometimes comparable with the strictly magnetic inelastic scattering [R. D. Lowde, Proc. Roy. Soc. (London) A235, 305 (1956)] given by γ'. In the region of the critical point $\langle S^z\rangle$ is small and the scattering from γ' large, so this experimental difficulty will not arise. However, if measurements of energy transfer are made it may be possible to observe magnetovibrational effects since the magnetic scattering from γ' is nearly elastic. The major magnetovibrational term is now, however, likely to arise from γ' which is giving the largest scattering in this temperature region.

[17] U. Firgau, Ann. Physik 40, 295 (1941).

by taking independent clusters of spins comprising one spin and all its neighbors. The interaction of this cluster with the rest of the crystal is assumed to take the form of a magnetic field H_1 which acts on the outer atoms of the cluster. If $H_1 \neq 0$ the system is ordered and the degree of order may be conveniently discussed in terms of an ordering parameter

$$\lambda = \exp(-2\beta H_1/kT). \tag{3.2}$$

Taking a single cluster with effective Hamiltonian,

$$\mathcal{H} = \sum_{\beta} -2JS^z(\mathbf{R})S^z(\mathbf{R}+\boldsymbol{\beta}) - 2\beta H_1 S^z(\mathbf{R}+\boldsymbol{\beta}), \tag{3.3}$$

the probability that the central atom $\mathbf{R}$ has $S^z = +\frac{1}{2}$

$$P^+(\mathbf{R}) = (1+\lambda t)^z/(1+\lambda t)^z + (\lambda+t)^z, \tag{3.4}$$

while for

$$S^z = -\tfrac{1}{2}, \quad P^-(\mathbf{R}) = (\lambda+t)^z/(1+\lambda t)^z + (\lambda+t)^z \tag{3.5}$$

where $t = e^{-J/kT}$. For an atom on the outside of the cluster, however,

$$\begin{aligned} P^+(\mathbf{R}+\boldsymbol{\beta}) = (1+\lambda)^{z-1} \\ + t(t+\lambda)^{z-1}/(1+t\lambda)^z + (t+\lambda)^z. \end{aligned} \tag{3.6}$$

But the probability that a spin has $S^z = +\frac{1}{2}$ must be the same whether it be regarded as at the center or on the outside of a cluster. Hence, (3.4) and (3.6) must be equal, giving

$$\lambda(1+t\lambda)^{z-1} = (t+\lambda)^{z-1} \tag{3.7}$$

as an equation for λ. This has only one root $\lambda = 1$ (i.e., no order) above a certain temperature given by

$$t = z - 2/z,$$

i.e.,

$$kT_c = J\left(\log\frac{z}{z-2}\right)^{-1}. \tag{3.8}$$

Below this critical temperature (3.7) has a solution which can be shown to give a lower free energy than the solution $\lambda = 1$, and so describes an ordered state. Using (3.4), (3.5), and (3.7)

$$\langle S^z \rangle = (1-\lambda^2)/2(1+2t\lambda+\lambda^2). \tag{3.9}$$

The quantity $\gamma^{zz}(\mathbf{R},0)$ may be written

$$\begin{aligned} \gamma(\mathbf{R}) = \langle S_0^z S^z(\mathbf{R}) \rangle &= \gamma'(\mathbf{R}) + \langle S^z \rangle^2 \\ &= \tfrac{1}{4}[P^{++}(0,\mathbf{R}) + P^{--}(0,\mathbf{R}) \\ &\quad - P^{+-}(0,\mathbf{R}) - P^{-+}(0,\mathbf{R})], \end{aligned} \tag{3.10}$$

where $P^{++}(0,\mathbf{R})$ is the probability that the spin at 0 be + if the spin at $\mathbf{R}$ be +, etc. We now calculate $P^{++}(0,\mathbf{R})$ by considering the clusters which include $\mathbf{R}$.

The fact that the spin at zero is fixed as + will affect these clusters, since it will exert a force through the coupling betweeni ntermediate atoms and makes $P^+(\mathbf{R})$ greater than its equilibrium value. The effect of the environment of the cluster is transmitted through the effective magnetic field. Thus the fixing of spin zero can be considered as modifying H_1. At large distances $\mathbf{R}'$ this modification will be small. λ must therefore be modified and replaced by $\lambda + \epsilon_\alpha(\mathbf{R}')$ ($\epsilon \ll \lambda$). Here α labels the atoms on the outside of the cluster centered at $\mathbf{R}'$ and ϵ will vary with α and $\mathbf{R}'$ since it varies with the distance from 0.

$P^{++}(0, \mathbf{R}+\boldsymbol{\beta})$ can be calculated in two ways in parallel to (3.5) and (3.6). For spin $\mathbf{R}$ considered at the center of a cluster

$$P^{++}(0,\mathbf{R}) = \frac{P^+(0) \prod_\beta (1+t\lambda+t\epsilon_\beta(\mathbf{R}))}{\prod_\beta (1+t\lambda+t\epsilon_\beta(\mathbf{R})) + \prod_\beta (t+\epsilon_\beta(\mathbf{R})+\lambda)}, \tag{3.11}$$

where $\prod_\beta$ is the product over the z neighbors. Writing

$$\sigma(\mathbf{R}') = \sum_\alpha \epsilon_\alpha(\mathbf{R}'), \tag{3.12}$$

where the sum is over the neighbors of $\mathbf{R}'$, and using (3.7), the analog of (3.11) for $\mathbf{R}+\boldsymbol{\beta}$ regarded as the center of a cluster is, to first order in ϵ

$$P^{++}(0, \mathbf{R}+\boldsymbol{\beta}) = \frac{(1+t\lambda)^2}{(1+\lambda^2+2t\lambda)^2}\left[1 - \frac{(1-t^2)\sigma(\mathbf{R}+\boldsymbol{\beta})}{(1+\lambda^2+2t\lambda)(1+t\lambda)}\right]. \tag{3.13}$$

Alternatively considering $\mathbf{R}+\boldsymbol{\beta}$ as a spin on the outside of the cluster around $\mathbf{R}$,

$$P^{++}(0, \mathbf{R}+\boldsymbol{\beta}) = \frac{P^{++}(0,\mathbf{R})}{1+t\lambda+t\epsilon_\beta(\mathbf{R})} + \frac{tP^{+-}(0,\mathbf{R})}{t+\lambda+\epsilon_\beta(\mathbf{R})}. \tag{3.14}$$

Equating (3.13) and (3.14), the terms independent of ϵ vanish and the first-order terms give

$$\begin{aligned} \sigma(\mathbf{R}+\boldsymbol{\beta}) = \frac{\lambda(1-t^2)}{(t+\lambda)(1+t\lambda)}\sigma(\mathbf{R}) \\ + \epsilon_\beta(\mathbf{R})\frac{t(1+2\lambda t+\lambda^2)(t+t\lambda^2+2\lambda)}{\lambda(t+\lambda)(1+t\lambda)(1-t^2)} \cdots. \end{aligned} \tag{3.15}$$

Summing over $\boldsymbol{\beta}$, the neighbors of $\mathbf{R}$, and using (3.12) we have

$$\begin{aligned} \sum_\beta \sigma(\mathbf{R}+\boldsymbol{\beta}) = \sigma(\mathbf{R})\biggl[\frac{z\lambda(1-t^2)}{(t+\lambda)(1+t\lambda)} \\ + \frac{t(1+2\lambda t+\lambda^2)(t+t\lambda^2+2\lambda)}{(t+\lambda)(1+t\lambda)(1-t^2)}\biggr]. \end{aligned} \tag{3.16}$$

A similar calculation of $P^{+-}(0,\mathbf{R})$ shows that the first-order changes are as above proportional to $\sigma(\mathbf{R})$ which is governed by (3.16). The behavior of $P^{-+}(0,\mathbf{R})$ and $P^{--}(0,\mathbf{R})$ can be calculated in the same way. By setting the spin at 0 as −, λ will be modified to become $\lambda + \epsilon_\alpha'(\mathbf{R})$ on the cluster around $\mathbf{R}'$. Since the prob-

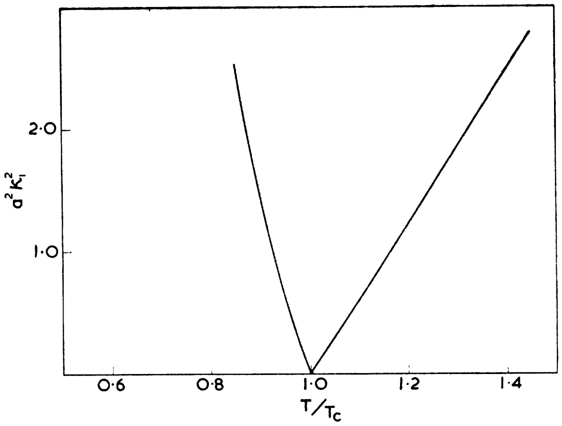

FIG. 1. $a^2\kappa_1^2$ for the Ising model, spin $\frac{1}{2}$.

ability of any atom being + is the same,

$$\begin{aligned}P^{+-}(0,\mathbf{R})+P^{++}(0,\mathbf{R})&=P^{-+}(0,\mathbf{R})+P^{++}(0,\mathbf{R})\\&=P^{-+}(0,\,\mathbf{R}+\boldsymbol{\beta})+P^{++}(0,\,\mathbf{R}+\boldsymbol{\beta})\\&=(1+t\lambda)/(1+\lambda^2+2\lambda t),\end{aligned}\tag{3.17}$$

and one can show after some manipulation that to first order in ϵ

$$(1+t\lambda)\epsilon_\alpha(\mathbf{R}')+\lambda(\lambda+t)\epsilon_\alpha'(\mathbf{R}')=0\tag{3.18}$$

everywhere. Each $P^{ij}(0,\mathbf{R})$ therefore can be obtained by modifying (3.13).

The first-order changes in $P^{ij}(0,\mathbf{R})$ are all proportional to $\sigma(\mathbf{R})$ and hence from (3.10) so is $\gamma'(\mathbf{R})$. Modifying (3.16) we obtain a difference equation for γ', *viz.*,

$$\sum_\beta \gamma'(\mathbf{R}+\boldsymbol{\beta})-z\gamma'(\mathbf{R})-a^2\kappa_1^2\gamma'(\mathbf{R})=0,\tag{3.19}$$

where

$$a^2\kappa_1^2=\frac{t(1+\lambda^2+2\lambda t)}{(t+\lambda)(1+\lambda t)}\left[\frac{(t+t\lambda^2+2\lambda)}{\lambda(1-t^2)}-z\right].\tag{3.20}$$

At large distances $\mathbf{R}$ where γ' will be slowly varying (3.19) can be replaced by a differential equation

$$(\nabla^2-\kappa_1^2)\gamma'(\mathbf{R})=0\tag{3.21}$$

which was derived by Van Hove.[10] However, κ_1 has now been determined on a microscopic model. The solution is

$$\frac{V_0}{16\pi r_1^2 R}e^{-\kappa_1 R},\tag{3.22}$$

where r_1 is an arbitrary constant which must be determined from the boundary conditions. Thus the effective range of correlation, κ_1^{-1}, becomes infinite as the Curie temperature is approached from either side. $a^2\kappa_1^2$ is plotted in Fig. 1 and is roughly linear with $|T-T_c|/T_c$ but varies twice as rapidly below as above T_c. The range of correlation becomes of order a when $|T-T_c|/T_c\sim 0.1$ and the slowly varying condition is broken down. Thus (3.22) is only valid over a narrow region about T_c. However, it is more convenient to work directly with the difference equation which has a much wider range of validity.

The long range of correlation near T_c means that if a particular atom has, say, a + spin, there is likely to be around it a region where spins are more often + than −. The equality in numbers of + and − spins is maintained by the presence elsewhere of regions of predominantly − spins. The size of these regions is $\sim\kappa^{-1}$ and becomes macroscopic at T_c itself. These extended regions are the fluctuations that cause the increased scattering; the neutron effectively seeing many changes of refractive index as it traverses the crystal. Because of their size the scattering is predominantly small angle.

To calculate the scattering cross section as the Fourier transform of $\gamma'(\mathbf{R})$ it is essential to consider the behavior of $\gamma'(\mathbf{R})$ at small $\mathbf{R}$ where (3.19) is no longer valid and the approximate solution (3.22) diverges. This divergence, if included, greatly increases the scattering at large $\boldsymbol{\kappa}$ so that Van Hove's cross section obtained in this way [Eq. (41) of reference (10)] actually diverges when summed over reciprocal lattice points $\boldsymbol{\tau}$. (This is not saved by the Debye-Waller factor which should read $\exp-\langle(\boldsymbol{\kappa}\cdot\mathbf{u})^2\rangle_T$.) Attempts to avoid this difficulty by arbitrarily cutting off (3.22) at some small R and replacing it by calculated values of $\gamma'(\mathbf{R})$ at small distances can lead to negative values of the cross section—again a spurious effect. This behavior is not surprising, since the conditions necessary to produce positive definite transforms are known to be stringent in the theory of continuous functions.‡

These difficulties may be overcome by working with the difference equation itself, when the cross section can be obtained in a simple form. Equation (3.19) was an approximation valid at large $\mathbf{R}$ and we may suppose that the equation for $\gamma'(\mathbf{R})$ may be written

$$\textstyle\sum_\beta \gamma'(\mathbf{R}+\boldsymbol{\beta})-z\gamma'(\mathbf{R})-a^2\kappa_1^2\gamma'(\mathbf{R})=-p(\mathbf{R})\tag{3.23}$$

if $p(\mathbf{R})$ is suitably defined. At large $\mathbf{R}$, as discussed in the argument leading to (3.16), $p(\mathbf{R})$ is zero to first order in ϵ and hence in $\gamma'(\mathbf{R})$. By a straightforward but tedious extension of that discussion it is found that $p(\mathbf{R})$ is actually of order ϵ^3. This argument may be briefly summarized when $T>T_c$ so that $\lambda=1$. The consistency equation (3.17) is satisfied identically by

$$\epsilon_\alpha'(\mathbf{R}')=-\epsilon_\alpha(\mathbf{R})/[1+\epsilon_\alpha(\mathbf{R})],\tag{3.24}$$

and to third order in ϵ

$$\begin{aligned}\gamma'(\mathbf{R})=-\frac{(1-t)}{8(1+t)}\Bigg\{&\sigma(\mathbf{R})-\tfrac{1}{2}\eta(\mathbf{R})\\&-\frac{(1-t)^2}{12(1+t)^2}\left[\sigma(\mathbf{R})^3-\frac{4(1-t^3)}{(1-t)^3}\xi(\mathbf{R})\right]\Bigg\},\end{aligned}\tag{3.25}$$

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‡ See, for example, Cramer's *Mathematical Methods of Statistics* (Princeton University Press, Princeton, 1946), p. 91.

where

$$\eta(\mathbf{R})=\sum_\alpha \epsilon_\alpha(\mathbf{R})^2, \quad \xi(\mathbf{R})=\sum_\alpha \epsilon_\alpha(\mathbf{R})^3. \qquad (3.26)$$

By calculating $P^{++}(0,\mathbf{R})$, etc., in two ways as before, but to third order, the equation (3.23) is found to hold if

$$p(\mathbf{R})=\frac{t(1-t)^2}{12(1+t)^4}\{\xi(\mathbf{R})-\tfrac{3}{2}\sigma(\mathbf{R})\eta(\mathbf{R})+\tfrac{1}{2}\sigma(\mathbf{R})^3\}. \qquad (3.27)$$

Thus at large distances $p(\mathbf{R})\sim\epsilon(\mathbf{R})^3$. It is sufficient to evaluate (3.27) using the first-order expressions (3.15) for ϵ and (3.16) for σ and the approximate solution (3.22). Then using (3.26)

$$p(\mathbf{R})=-\frac{3}{8}\left[\frac{V_0}{4\pi r_1^2}\frac{e^{-\kappa_1 R}}{R}\right]^3 \qquad (3.28)$$

apart from terms smaller by a factor $a^2\kappa_1^2$ which may be neglected near T_c. It is therefore a good approximation to take $p(\mathbf{R})=0$ if $\kappa_1 R>1$, although the range over which this appears to be bad increases as $\kappa_1\rightarrow 0$ near T_c. It is then necessary to consider higher order terms in the expansion in ϵ.

However, at the first few lattice spacings $p(\mathbf{R})$ may be calculated directly from a single cluster. One finds

$$\gamma_0'=\lambda(t+\lambda)(1+t\lambda)/(1+\lambda^2+2t\lambda)^2 \qquad (3.29)$$

for nearest neighbors

$$\gamma_1'=\lambda^2(1-t^2)/(1+\lambda^2+2t\lambda)^2, \qquad (3.30)$$

and for all other pairs in a cluster irrespective of their relative positions

$$\gamma'=\lambda^3(1-t^2)/(1+\lambda^2+2t\lambda)^2(t+\lambda)(1+t\lambda). \qquad (3.31)$$

For (3.23) therefore

$$p_0=t(t+t\lambda^2+2\lambda)/(1-t^2)(1+\lambda^2+2t\lambda), \qquad (3.32)$$

$$p_1=0. \qquad (3.33)$$

The solution of the difference equation (3.23) is simply obtained in terms of the Fourier transform

$$\phi(\mathbf{t})=\sum_\mathbf{R} \gamma'(\mathbf{R}) \exp(i\mathbf{t}\cdot\mathbf{R}). \qquad (3.34)$$

Summing over the equations with weight $\exp(i\mathbf{t}\cdot\mathbf{R})$

$$\phi(\mathbf{t})=\frac{\sum_\mathbf{R} p(\mathbf{R}) \exp(i\mathbf{t}\cdot\mathbf{R})}{\sum_\beta (1-\exp(i\mathbf{t}\cdot\boldsymbol{\beta})+a^2\kappa_1^2}. \qquad (3.35)$$

Some further information about the p's can be obtained from the relation of $\phi(0)$ to the susceptibility $\chi=(\partial M/\partial H)_{H=0}$, where M is the magnetic moment of the system

$$\begin{aligned}\phi(0)&=\sum_\mathbf{R} \gamma'(\mathbf{R})=\sum_\mathbf{R} \langle S_0^zS^z(\mathbf{R})\rangle-N\langle S_0^z\rangle^2\\ &=N^{-1}\langle\sum_\mathbf{R} S^z(\mathbf{R})\rangle^2-(N+1)\langle S_0^z\rangle^2\\ &=\frac{1}{N}\left[\left(\frac{M}{2\beta}\right)^2+\frac{kT\chi}{4\beta^2}-\left(\frac{M}{2\beta}\right)^2\right]=\frac{kT\chi}{4N\beta^2}. \qquad (3.36)\end{aligned}$$

The calculation of χ on the B-P theory is well known[17] and using this with (3.35) and (3.20) one finds

$$\sum_\mathbf{R} p(\mathbf{R})=p_0. \qquad (3.37)$$

From this result and the particular $p_\mathbf{R}$ of (3.28), (3.32), and (3.33) it is seen to be a good approximation for (3.23) to let

$$p(\mathbf{R})=0, \quad \mathbf{R}\neq 0, \qquad (3.38)$$

while p_0 is given by (3.32). We use this approximation very close to T_c when (3.28) is not valid.

The Fourier transform of γ' has then a simple form

$$\phi(\mathbf{t})=\frac{p_0}{\sum_\beta (1-\exp(i\mathbf{t}\cdot\boldsymbol{\beta}))+a^2\kappa_1^2}, \qquad (3.39)$$

and so has the cross section (2.2) with static approximation (2.7),

$$\frac{d^2\sigma}{d\Omega dE}=\left(\frac{2ge^2}{mc^2}\right)^2\frac{1}{h}\frac{k}{k_0}|F(\boldsymbol{\kappa})|^2e^{-2W}(1-e_z^2)$$

$$\times\delta(\omega)\frac{p_0}{\sum_\beta (1-\exp(i\mathbf{t}\cdot\boldsymbol{\beta}))+a^2\kappa_1^2}. \qquad (3.40)$$

The factor in the cross section arising from ϕ is periodic in reciprocal space because of the periodicity in the lattice of $\mathbf{R}$, and as may be seen in (3.39). The scattering is peaked around the Bragg directions when $\boldsymbol{\kappa}=\boldsymbol{\tau}$ and falls off at large $\boldsymbol{\tau}$ because of the form factor and the Debye-Waller factor.

For the two lattices considered

$$\sum_\beta(1-\exp(i\boldsymbol{\kappa}\cdot\boldsymbol{\beta}))=6-2\cos\kappa_xa-2\cos\kappa_ya-2\cos\kappa_za \quad \text{for s.c.}$$

and (3.41)

$$=8(1-\cos\tfrac{1}{2}\kappa_xa\cos\tfrac{1}{2}\kappa_ya\cos\tfrac{1}{2}\kappa_za) \quad \text{for b.c.c.}$$

For small $\boldsymbol{\kappa}-\boldsymbol{\tau}$ both these expressions are approximately

$$=a^2|\boldsymbol{\kappa}-\boldsymbol{\tau}|^2. \qquad (3.42)$$

Thus the cross section close to reciprocal lattice points is given by the principal term in the sum over $\boldsymbol{\tau}$ given by Van Hove.[10] The factor p_0 is related to the second length r_1 introduced as in (3.22) by

$$p_0=a^2/4r_1^2. \qquad (3.43)$$

The parameters κ_1, r_1, defining the scattering as given by (3.20) and (3.32) are plotted in Figs. 1 and 2 for the b.c.c. lattice. Just above T_c,

$$a^2\kappa_1^2=\frac{(z-2)^2z}{2(z-1)}\ln\left(\frac{z}{z-2}\right)\frac{T-T_c}{T_c}, \qquad [(3.44)$$

and the slope is just twice as great below T_c. Thus, r_1 is a monatomic decreasing function with increasing T.

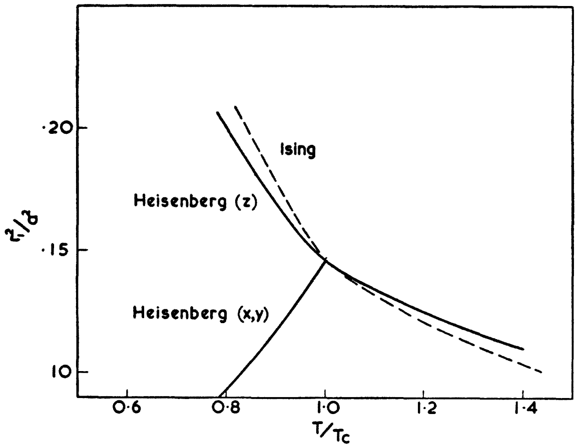

FIG. 2. r_1^2/a^2 for the Ising and Heisenberg models, spin $\frac{1}{2}$.

At T_c itself

$$r_1^2/a^2=(z-1)/z(z-2). \tag{3.45}$$

Although the cross section has this simple form, γ' is actually a rather complicated function. As the transform of ϕ it is

$$\gamma'(\mathbf{R})=\frac{V_0}{(2\pi)^3}\int_{\text{cell}}\phi(t)\exp(-i\mathbf{t}\cdot\mathbf{R})dt, \tag{3.46}$$

where the integral is over a unit cell in reciprocal space. At large $\mathbf{R}$ the approximate solution (3.22) is reproduced by using (3.42) and (3.43) and extending the integration to all space. Slater and Koster[18] give a solution of the difference equation in the simple cubic lattice as

$$\gamma'(\mathbf{R})=\tfrac{1}{2}p\gamma'(0)\int_0^\infty \exp[-(z+a^2\kappa_1^2)]t \times I_p(t)I_q(t)I_r(t)dt, \tag{3.47}$$

where p, q, r are the components of $\mathbf{R}$ in units of a and I is a Bessel function of imaginary argument. Slater and Koster have computed γ' with the exponent in (3.34) equal to 3.5 which is approximately half the values of interest in this problem. They find that an approximate formula like (3.20) is reasonably accurate even at the smallest distances (see their Fig. 3) but that the crystal structure gives appreciable directional asymmetry as might be expected from (3.31).

Calculations of κ_1 and r_1 can also be made from Zernike's results[13] and these differ from the above by only a few percent close to T_c. The methods are in fact very similar in this temperature region although Zernike neglected correlation between two spins on the periphery of a cluster and the effect of the ordering field H_1. His results, moreover, do not come in such a simple form and our method may be readily extended to other cases which are considered in later sections.

4. HEISENBERG MODEL $S=\frac{1}{2}$

The direct extension of the Bethe-Peierls method to the Heisenberg model of a ferromagnet was made by Weiss[19] who calculated the detailed properties of a cluster with an effective Hamiltonian like (3.3) except that the Ising interaction (3.1) is replaced by the Heisenberg form

$$-2J\mathbf{S}(\mathbf{R})\cdot\mathbf{S}(\mathbf{R}+\boldsymbol{\rho}). \tag{4.1}$$

The calculations are however much more complicated and require numerical analysis, since the eigenstates are made more difficult by the noncommutation properties of $\mathbf{S}(\mathbf{R})$. It is possible to extend the theory of the last section into this formalism but numerical solutions are again required.

Recently Van Kranendonk and Kasteleijn[20] have given a different formulation called the constant coupling approximation which allows the results of this model to be much more easily obtained in closed algebraic form in close analogy to the results of the last section. This is an extension of the so-called quasi-chemical approximation of Guggenheim[21] for the Ising problem. This approach chooses a basic cluster of a pair of spins. Returning for a moment to the Ising case the effective Hamiltonian is

$$-2JS^z(\mathbf{R})S^z(\mathbf{R}+\boldsymbol{\rho})-2\beta H_1(S^z(\mathbf{R})+S^z(\mathbf{R}+\boldsymbol{\rho})), \tag{4.2}$$

the effective field H_1 again giving the effects of the surroundings. H_1 is now determined by minimizing the free energy. The resultant equation is found to be (3.7) with the same definition of λ(3.2). In fact the two methods are exactly equivalent as was shown by Fowler and Guggenheim.[22] For the probabilities of occurrence of the various configurations of nearest neighbors can be found by inspection of (4.2)

$$P^{++}:P^{--}:P^{+-}:P^{-+}=1/\lambda:\lambda:t:t, \tag{4.3}$$

and they are found to be the same in the B-P method of the last section when the condition (3.7) is used.

Taking now (4.2) with the Heisenberg interaction (4.1) substituted, the eigenstates and eigenvalues are

$$\begin{aligned} &|++\rangle; && -\tfrac{1}{2}J-2\beta H_1\\ &\frac{1}{\sqrt{2}}[|+-\rangle+|-+\rangle]; && -\tfrac{1}{2}J\\ &|--\rangle; && -\tfrac{1}{2}J+2\beta H_1\\ &\frac{1}{\sqrt{2}}[|+-\rangle-|-+\rangle]; && \tfrac{3}{2}J \end{aligned} \tag{4.4}$$

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[18] J. C. Slater and G. F. Koster, Phys. Rev. **96**, 1208 (1954).

[19] P. R. Weiss, Phys. Rev. **74**, 1493 (1948).

[20] J. Van Kranendonk and P. W. Kasteleijn, Physica **22**, 317 (1956).

[21] E. A. Guggenheim, Proc. Roy. Soc. (London) **A148**, 304 (1935).

[22] R. H. Fowler and E. A. Guggenheim, Proc. Roy. Soc. (London) **A174**, 189 (1940).

and

$$P^{++}:P^{--}:P^{+-}:P^{-+}=\frac{1}{\lambda}:\lambda:\tfrac{1}{2}(1+t^2):\tfrac{1}{2}(1+t^2). \quad (4.5)$$

Comparing this with (4.3) the properties of this model are seen to be exactly the same as the Ising model with $\frac{1}{2}(1+t^2)$ replacing t, as is shown by detailed calculation in reference 20.

The two methods are, however, not equivalent for the Heisenberg model. This arises because the noncommuting properties of $\mathbf{S}(\mathbf{R})$ have been partly neglected in the constant coupling approximation, and enter only through the effective field. At high spin values where this effect is less important Van Kranendonk and Kasteleijn show that their results are almost identical with the results of the Weiss theory obtained by Brown and Luttinger.[23] It is not easy to see *a priori* which approximation is more correct since the effective field enters differently into the two theories and may in part compensate for the neglect of commutation in reference 20. In fact these authors show that their method is better at high temperatures in the sense that it reproduces the exact high-temperature expansion of the susceptibility to one further term than the Weiss method.

For the Heisenberg model the $\gamma'^{\alpha\beta}(\mathbf{R})$ are required for all components α, β of the spins. By symmetry it is clear that $\gamma'^{\alpha\beta}(\mathbf{R})=0$ if $\alpha\neq\beta$. If the direction of spin in the ordered state is taken as z then $\gamma^{zz}(\mathbf{R})\neq\gamma^{xx}(\mathbf{R})=\gamma^{yy}(\mathbf{R})$ and there are essentially two different correlation functions to be determined. Above T_c however the system is isotropic and $\gamma^{zz}(\mathbf{R})=\gamma^{xx}(\mathbf{R})$.

To obtain these functions of position it is convenient to construct a theory of the B-P kind and use an argument like that of Sec. 3. We therefore build a cluster theory equivalent to the constant coupling approximation. This is not difficult in view of the close analogy with the Ising case discussed above. Considering a spin $\mathbf{R}$ as participating in z pairs and using (4.5) the probabilities of its being + and − have the form

$$\begin{aligned} P^{+}&\sim(P^{++}+P^{+-})^{z}=\left[\frac{1}{\lambda}+\tfrac{1}{2}(1+t^2)\right]^{z}, \\ P^{-}&\sim(P^{-+}+P^{--})^{z}=[\lambda+\tfrac{1}{2}(1+t^2)]^{z}. \end{aligned} \quad (4.6)$$

To obtain the relative probabilities we must take account of the fact that in the pair formulation an ordering field acts on spin $\mathbf{R}$, but its effect must not be included in the cluster where all the neighbors of $\mathbf{R}$ are considered. We therefore write

$$P^{+}:P^{-}=\nu^{z}\left(\frac{1}{\lambda}+\tfrac{1}{2}(1+t^2)\right)^{z}:(\lambda+\tfrac{1}{2}(1+t^2))^{z}, \quad (4.7)$$

where ν gives the effect of the field. By constructing the consistency equation for this cluster in the B-P manner and comparing it with the equation of the constant coupling approximation we find $\nu=\lambda=e^{-2\beta H_1/kT}$ as might reasonably be expected on physical grounds.

Here, $\gamma'^{zz}(\mathbf{R})$ is calculated as before from $P^{ij}(0,\mathbf{R})$. The fixed orientation of the spin at zero is considered to modify the effective fields acting on the pairs around $\mathbf{R}$ by inducing additional components $h\beta^{z}(\mathbf{R})$ in the z direction. Under these conditions the probabilities of spin arrangement in the pair $\mathbf{R}$, $\mathbf{R}+\boldsymbol{\beta}$ are given by (4.5) with λ modified.

$$\begin{aligned} P^{++}:P^{--}:P^{+-}:P^{-+}=1/(\lambda+\epsilon_{\beta}(\mathbf{R})) \\ :\lambda+\epsilon_{\beta}(\mathbf{R}):\tfrac{1}{2}(1+t^2):\tfrac{1}{2}(1+t^2). \quad (4.8) \end{aligned}$$

The properties of the spin at $\mathbf{R}$ must be obtained from the z pairs in which it participates which are no longer equivalent. So the cluster around $\mathbf{R}$ gives from (4.7)

$$\begin{aligned} P^{i+}(0,\mathbf{R}):P^{i-}(0,\mathbf{R})=\prod_{\beta}\left[(\lambda+\epsilon_{\beta}{}^{i}(\mathbf{R}))\tfrac{1}{2}(1+t^2)+1\right] \\ :\prod_{\beta}\left[\lambda+\epsilon_{\beta}{}^{i}(\mathbf{R})+\tfrac{1}{2}(1+t^2)\right]. \quad (4.9) \end{aligned}$$

The calculation therefore proceeds exactly as in Sec. 3 with $\frac{1}{2}(1+t^2)$ substituted for t. The Fourier transform of $\gamma'^{zz}(\mathbf{R})$ is then given by (3.39)

$$\phi^{zz}(\mathbf{t})=\frac{p_0(z)}{\sum_{\beta}(1-\exp(i\mathbf{t}\cdot\boldsymbol{\beta}))+a^2\kappa_{1z}{}^2}, \quad (4.10)$$

where

$$p_0(z)=\frac{a^2}{4r_{1z}{}^2}=\frac{(1+t^2)[(1+t^2)(1+\lambda^2)+4\lambda]}{(1-t^2)(3+t^2)(1+\lambda^2+\lambda(1+t^2))} \quad (4.11)$$

$$\begin{aligned} a^2\kappa_{1z}{}^2=&\frac{2(1+t^2)[1+\lambda^2+\lambda(1+t^2)]}{(1+t^2+2\lambda)[2+\lambda(1+t^2)]} \\ &\times\left\{\frac{2[(1+t^2)(1+\lambda^2)+4\lambda]}{\lambda(1-t^2)(3+t^2)}-z\right\}. \quad (4.12) \end{aligned}$$

These parameters are plotted and compared with the Ising model in Figs. 2 and 3.

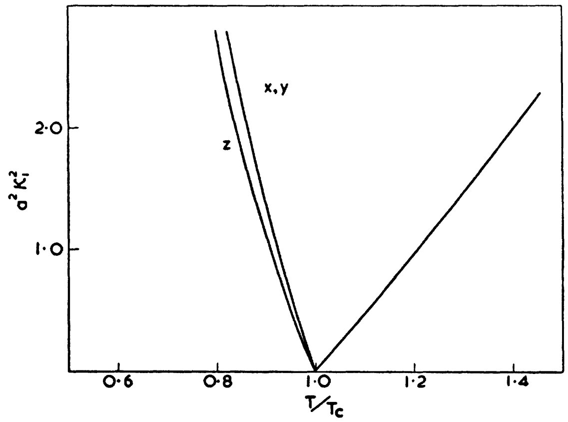

FIG. 3. $a^2\kappa_1{}^2$ for the Heisenberg model, spin $\frac{1}{2}$.

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[23] H. A. Brown and J. M. Luttinger, Phys. Rev. 100, 685 (1955).

For γ'^{xx} we consider a cluster at $\mathbf{R}$ and take the small additional effective field $h_\beta{}^x(\mathbf{R})$ from the fixing of the component of the spin at 0 in the x direction. Thus the effective Hamiltonian for a pair is

$$-2J\mathbf{S}(\mathbf{R})\cdot\mathbf{S}(\mathbf{R}+\boldsymbol{\beta})-2\beta H_1(\mathbf{S}^z(\mathbf{R})+\mathbf{S}^z(\mathbf{R}+\boldsymbol{\beta})) \\ -2\beta h_\beta{}^x(\mathbf{R})(\mathbf{S}^x(\mathbf{R})+\mathbf{S}^x(\mathbf{R}+\boldsymbol{\beta})). \qquad (4.13)$$

The total field

$$H=(H_1{}^2+h_\beta{}^x(\mathbf{R})^2)^{\frac{1}{2}} \qquad (4.14)$$

acts in a direction θ to x where $\tan\theta=h_\beta{}^x/H_1$. Referred to this direction the eigenfunctions have the form (4.4) and the energies are the same with H replacing H_1. Changing the axis of reference to x these functions transform to

$$\begin{aligned}(1,1)'&=|++\rangle=\tfrac{1}{2}(1+\sin\theta)(1,1) \\ &\quad+\tfrac{1}{2}(1-\sin\theta)(1,-1)+\frac{1}{\surd 2}\cos\theta(1,0) \\ (1,0)'&=\frac{1}{\surd 2}[|+-\rangle+|-+\rangle] \\ &=\frac{1}{\surd 2}\cos\theta[(1,1)-(1,-1)]+\sin\theta(1,0) \qquad (4.15) \\ (1,-1)'&=|--\rangle=\tfrac{1}{2}(1-\sin\theta)(1,1) \\ &\quad+\tfrac{1}{2}(1+\sin\theta)(1,-1)+\frac{1}{\surd 2}\cos\theta(1,0) \\ (0,0)'&=\frac{1}{\surd 2}[|+-\rangle-|-+\rangle]=(0,0),\end{aligned}$$

where the representation in parentheses gives the total spin and its component referred to the axis of H on the left and to x on the right. The coefficients are the elements $\mathfrak{D}^{(1)}mm'$ of the matrix irreducible representation of the rotation group. Defining $\lambda=e^{-2\beta H}/kT$ we have from (4.15)

$$\begin{aligned}&P_{(x)}{}^{++}:P_{(x)}{}^{--}:P_{(x)}{}^{+-}:P_{(x)}{}^{-+} \\ &\quad=(\lambda+1)^2+2(\lambda^2-1)\sin\theta+(\lambda-1)^2\sin^2\theta \\ &\qquad:(\lambda+1)^2-2(\lambda^2-1)\sin\theta+(\lambda-1)^2\sin^2\theta \\ &\qquad:(\lambda^2+1+2\lambda t^2)+(\lambda-1)^2\sin^2\theta \\ &\qquad\quad:(\lambda^2+1+2\lambda t^2)+(\lambda-1)^2\sin^2\theta. \qquad (4.16)\end{aligned}$$

Since h^x is always small $(\lambda-1)\sin\theta$ is small. Well below T_c, $\sin\theta$ is always small $\sim h^x/H_1$, and when H_1 itself becomes small close to T_c, $\lambda-1$ is small $\sim 2\beta H/kT_c$, so that $(\lambda-1)\sin\theta\sim 2\beta h^x/kT_c$. To first order in h^x therefore $(\lambda-1)^2\sin^2\theta$ may be neglected and (4.16) written

$$\begin{aligned}&P_{(x)}{}^{++}:P_{(x)}{}^{--}:P_{(x)}{}^{+-}:P_{(x)}{}^{-+} \\ &\quad=(1-\epsilon):(1+\epsilon):(\lambda^2+1+2\lambda t^2)/(\lambda+1)^2 \\ &\qquad\quad:(\lambda^2+1+2\lambda t^2)/(\lambda+1)^2, \qquad (4.17)\end{aligned}$$

where ϵ is small and linear in h^x.

$\gamma'^{xx}(\mathbf{R})$ is calculated like $\gamma'^{zz}(\mathbf{R})$ by building an equi alent B-P theory for z pairs with properties defined l (4.17). Comparing (4.17) with (4.8) it is seen that (4.1 may be obtained by putting $\lambda=1$ and

$$\tfrac{1}{2}(1+t^2)\rightarrow(\lambda^2+1+2\lambda t^2)/(\lambda+1)^2 \qquad \text{(4.1}$$

in that expression. The argument of Sec. 3 may thei fore be repeated with (4.18) for t to give $\gamma^{xx}(\mathbf{R})$ and ¢ in the form (3.39) with

$$p_0(x)=\frac{a^2}{4r_{1x}{}^2}=\frac{(\lambda^2+1+2\lambda t^2)(\lambda+1)^2}{4\lambda^2(1-t^2)(\lambda^2+1+\lambda(1+t^2))} \qquad \text{(4.1}$$

$$a^2\kappa_{1x}{}^2=\frac{(\lambda^2+1+2\lambda t^2)}{(\lambda^2+1+\lambda(1+t^2))}\left[\frac{(\lambda+1)^2}{\lambda(1-t^2)}-z\right] \qquad \text{(4.2}$$

which are plotted in Figs. 2 and 3.

Substituting into Eq. (1.2) the cross section is in t static approximation

$$\frac{d^2\sigma}{d\Omega dE}=\left(\frac{2ge^2}{mc^2}\right)^2\frac{k}{hk_0}|F(\boldsymbol{\kappa})|^2e^{-2W}\delta(\omega) \\ \times\left[\frac{(1+e_z{}^2)p_0(x)}{\sum_\beta(1-\exp(i\mathbf{t}\cdot\boldsymbol{\beta}))+a^2\kappa_{1x}{}^2)}\right. \\ \left.+\frac{(1-e_z{}^2)p_0(z)}{\sum_\beta(1-\exp(i\mathbf{t}\cdot\boldsymbol{\beta}))+a^2\kappa_{1z}{}^2}\right]. \qquad \text{(4.2}$$

Below T_c therefore the cross section shows a depender on the direction of magnetization through e_z. Above this disappears as it must when both (4.11) and (4.1 become

$$\frac{a^2}{4r_1{}^2}=p_0=2(1+t^2)/(3+t^2)(1-t^2) \qquad \text{(4.2}$$

while (4.12) and (4.20) become

$$a^2\kappa_1{}^2=\frac{2(1+t^2)}{(3+t^2)}\left[\frac{4}{1-t^2}-z\right]. \qquad \text{(4.2}$$

Thus, $\kappa_{1x}{}^2$ is always larger than $\kappa_{1z}{}^2$ below T_c. Close T_c, $\kappa_{1z}{}^2$ rises twice as fast as $\kappa_1{}^2$ above T_c, $\kappa_{1x}{}^2$ rises times as fast. $r_1{}^2$ has the value (3.45) at T_c and $r_{1x}{}^2$ c creases with decreasing T while $r_{1z}{}^2$ increases. This lea to a complicated behavior of (4.21) with temperatu

Van Kranendonk and Kasteleijn[20] use their resu with $z=12$ to give a first approximation for the fa centered cubic lattice. To the same accuracy the resu of this section might be used for the same purpose.

5. BINARY ALLOYS

Simple binary alloys with equal numbers of A and atoms may conveniently be treated in the formalism the Ising model of Sec. 3, with some slight modificatio which in fact returns the theory to the form origina presented by Bethe. The A atoms are taken to correspond to spin state $S^z=+\frac{1}{2}$, B to $S^z=+\frac{1}{2}$. The energy of an alloy with respect to the energy of the pure A and B crystals can be written

$$E=\tfrac{1}{2}N_{AB}[V_{AA}+V_{BB}-2V_{AB}]=-\tfrac{1}{2}JN_{AB}, \qquad (5.1)$$

where N_{AB} is the number of AB pairs, and V_{AA} is the interaction energy between a pair of nearest neighbor A atoms, etc. For alloys which tend to form an ordered mixed phase $2V_{AB}<V_{AA}+V_{BB}$, i.e., $J>0$. The effective interaction in the Ising model may therefore be written

$$2JS^z(\mathbf{R})S^z(\mathbf{R}+\boldsymbol{\beta}), \qquad (5.2)$$

where the sign is different from (3.1), J remaining positive.

The ordered arrangement of the s.c. and b.c.c. lattices under consideration consists of two sublattices, one comprising mostly A atoms, the other mostly B. This effect can be represented in this method by introducing a "staggered" ordering field H_1 which acts in opposite directions on the two sublattices. Since in these lattices all z neighbors of a B site are on the A lattice and vice versa the effective Hamiltonian of a pair may be written

$$-2JS^z(\mathbf{R})S^z(\mathbf{R}+\boldsymbol{\beta})-2\beta H_1(S^z(\mathbf{R})-S^z(\mathbf{R}+\boldsymbol{\beta})), \qquad (5.3)$$

where $\mathbf{R}$ is on the A lattice, $\mathbf{R}+\boldsymbol{\beta}$ on the B lattice. There are two kinds of cluster in the B-P method depending on the sublattice of the central atom. However, these may be treated together by considering an atom be "right" (i.e., A atom on A sublattice, B on B) or "wrong" (B on A) instead of the probabilities of the occurrence of the two spin orientations. Using (5.3) the probabilities of the four possible configurations of a pair, writing superscripts R for right and W for wrong, are in the ratio

$$P^{RR}:P^{WW}:P^{RW}:P^{WR}=1/\lambda:\lambda:t:t, \qquad (5.4)$$

where λ and t are defined as in (3.2) and (3.5). In a cluster the probabilities that an atom be right or wrong are

$$P^R:P^W=(1+\lambda t)^z:(\lambda+t)^z. \qquad (5.5)$$

Thus all the expressions of Sec. 3 can hold if R is written for $+$ and W for $-$. Returning to the $\mathbf{S}^z$ notation, $\langle\mathbf{S}^z\rangle$ has different signs on the two sublattices.

$$\begin{aligned}\langle\mathbf{S}^z(A)\rangle=\tfrac{1}{2}[P^R-P^W]&=-\langle\mathbf{S}^z(B)\rangle\\ &=(1-\lambda^2)/2(1+\lambda^2+2t\lambda) \qquad (5.6)\end{aligned}$$

as in (3.9). Similarly,

$$\begin{aligned}\gamma(\mathbf{R})=\langle\mathbf{S}^z(0)\mathbf{S}^z(\mathbf{R})\rangle&=\gamma'(\mathbf{R})+\langle\mathbf{S}^z(0)\rangle\langle\mathbf{S}^z(\mathbf{R})\rangle\\ &=\tfrac{1}{4}\delta(\mathbf{R})[P^{RR}(0,\mathbf{R})+P^{WW}(0,\mathbf{R})\\ &\qquad -P^{RW}(0,\mathbf{R})-P^{WR}(0,\mathbf{R})], \qquad (5.7)\end{aligned}$$

where $\delta(\mathbf{R})$ is ± 1 according as whether $\mathbf{R}$ is on the same sublattice as 0 or not. The difference equation (3.23) which is derived using P^{RR}, etc., therefore takes the form

$$\textstyle\sum_\beta \gamma'(\mathbf{R}+\boldsymbol{\beta})+z\gamma'(\mathbf{R})+a^2\kappa_1{}^2\gamma'(\mathbf{R})=+p(\mathbf{R}) \qquad (5.8)$$

since all the neighbors of $\mathbf{R}$ lie on the other lattice; and the Fourier transform of the solution analogous to (3.39) is

$$\phi(t)=\frac{p_0}{\sum_\beta (1+\exp(i\mathbf{t}\cdot\boldsymbol{\beta}))+a^2\kappa_1{}^2}. \qquad (5.9)$$

In determining p_0 as from (3.36) the algebra is exactly the same as that case but because of the formulation is in terms of "right" and "wrong" the quantities do not have the same meaning. χ is, for example, not the chemical potential.

The neutron scattering takes place because of the interaction with the nuclei which can be represented by a Fermi pseudo-potential. There will be an incoherent part giving an isotropic background and a coherent part for which the two elements will have different scattering lengths a and b. The effective coherent interaction between a neutron at $\mathbf{r}$ and the crystal may be written therefore

$$\sum_{\mathbf{R}}\frac{h^2}{2m_0}[\tfrac{1}{2}(a+b)+(a-b)\mathbf{S}^z(\mathbf{R})]\delta(\mathbf{r}-\mathbf{R}). \qquad (5.10)$$

The first term gives Bragg scattering with the reciprocal lattice vectors of the lattice

$$\frac{d\sigma}{d\Omega}=\sum_\tau\frac{NV_0}{(2\pi)^3}\tfrac{1}{4}(a+b)^2e^{-2W}\delta(\boldsymbol{\kappa}-\boldsymbol{\tau}). \qquad (5.11)$$

The form factor is here unity because of the δ function interaction (5.10). The second term shows the effect of the ordering. Below the critical temperature where $\langle S^z\rangle\neq 0$ it also gives Bragg scattering but because $\langle S^z\rangle$ is different on the two sublattices it occurs at different angles. The cross section can conveniently be written

$$\frac{d\sigma}{d\Omega}=\sum_\tau\frac{NV_0}{(2\pi)^3}\tfrac{1}{4}(a-b)^2e^{-2W}\delta(\boldsymbol{\kappa}-\boldsymbol{\tau}-\mathbf{w}), \qquad (5.12)$$

where $\mathbf{w}$ is a vector in reciprocal space such that $\exp(i\mathbf{w}\cdot\mathbf{R})=\pm 1$ according as $\mathbf{R}$ is the distance between two atoms on the same or different sublattices.§ There is in addition the critical scattering given by

$$\begin{aligned}\frac{d^2\sigma}{d\Omega dE}&=\frac{(a-b)^2}{4h}\delta(\omega)e^{-2W}\sum_R\gamma'(\mathbf{R})\exp(i\boldsymbol{\kappa}\cdot\mathbf{R})\\ &=\frac{(a-b)^2}{4h}\delta(\omega)e^{-2W}\\ &\qquad\times\frac{p_0}{\sum_\beta(1+\exp(i\boldsymbol{\kappa}\cdot\boldsymbol{\beta}))+a^2\kappa_1{}^2} \qquad (5.13)\end{aligned}$$

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§ See R. J. Elliott and R. D. Lowde, Proc. Roy. Soc. (London) **A230**, 46 (1955). For s.c. one may choose $\mathbf{w}=\pi/a(1,1,1)$ for b.c.c., $2\pi/a(1,0,0)$.

from (5.9) where p_0 and $a^2\kappa_1^2$ are given by (3.32) and (3.20).

The denominator is periodic in reciprocal space and is peaked around the points $\boldsymbol{\tau}+\mathbf{w}$. $\sum_{\boldsymbol{\rho}}(1+\exp(i\boldsymbol{\kappa}\cdot\boldsymbol{\rho}))$ can be written $\sum_{\boldsymbol{\rho}}(1-\exp(i(\boldsymbol{\kappa}+\mathbf{w})\cdot\boldsymbol{\rho}))$ and the function has the forms given by (3.41) and (3.42) except for the addition of $\mathbf{w}$ to $\boldsymbol{\kappa}$. The critical scattering thus occurs around the superlattice Bragg reflections and does *not occur in the forward direction.* Within the approximation γ is independent of time since transitions between spin states here mean atomic displacements. The static approximation is therefore exact for this case and all the scattering is elastic as given in (5.13).

No observations have been reported of critical scattering in these substances. It should also be observable with x-rays where a formula like (5.13) will hold with appropriate modification of the interaction (5.10). An atomic form factor will appear in the cross section because of the size of the atomic charge cloud. One experimental difficulty will arise from the time required for a crystal to reach its equilibrium configuration.

6. ANTIFERROMAGNETS $S=\frac{1}{2}$

The methods of the earlier sections may be extended without difficulty to cover the case of antiferromagnetism arising from a Heisenberg interaction of opposite sign to that in Sec. 4. The cluster method for this case was examined by Li.[24] Kasteleijn and Van Kranendonk[25] have recently treated this problem in the constant coupling approximation and again find that the results can be obtained in closed algebraic form, although their actual values are not very different from Li's. We shall find their formalism convenient for the theory of critical scattering, and adopt their notation with the ordering field written H_1 instead of A_4. The algebra is somewhat heavy and since the method is exactly parallel with that of earlier sections, the discussion will be given in outline.

The effective Hamiltonian for a pair of spins is

$$2J\mathbf{S}(\mathbf{R})\cdot\mathbf{S}(\mathbf{R}+\boldsymbol{\rho})-2\beta H_1[S^z(\mathbf{R})-S^z(\mathbf{R}+\boldsymbol{\rho})], \tag{6.1}$$

where $\mathbf{R}$ is on the sublattice of predominantly + spin and $\mathbf{R}+\boldsymbol{\rho}$ on the other sublattice of predominantly − spin. J is again positive. The eigenstates and energies of (6.1) are

$$\begin{array}{ll}
|++\rangle & \tfrac{1}{2}J \\
|--\rangle & \tfrac{1}{2}J \\
\dfrac{1}{\sqrt{2}}\sin\dfrac{\omega}{2}[|+-\rangle+|-+\rangle]-\dfrac{1}{\sqrt{2}}\cos\dfrac{\omega}{2}[|+-\rangle-|-+\rangle] & \\
\qquad -\tfrac{1}{2}J-(J^2+4H_1^2\beta^2)^{\frac{1}{2}} & (6.2)\\
\dfrac{1}{\sqrt{2}}\cos\dfrac{\omega}{2}[|+-\rangle+|-+\rangle]+\dfrac{1}{\sqrt{2}}\sin\dfrac{\omega}{2}[|+-\rangle-|-+\rangle] & \\
\qquad -\tfrac{1}{2}J+(J^2+4H_1^2\beta^2)^{\frac{1}{2}} &
\end{array}$$

where $\sin\omega=2H_1\beta(J^2+4H_1^2\beta^2)^{-\frac{1}{2}}$. The probabilities of the occurrence of the various configuration of neighbors, using "right" and "wrong" as in Sec. 5, are

$$p^{RR}:p^{WW}:p^{RW}:p^{WR}=X_+:X_-:1:1, \tag{6.3}$$

where

$$X_\pm=\tfrac{1}{2}x\left[\left(v+\frac{1}{v}\right)\pm\sin\omega\left(v-\frac{1}{v}\right)\right] \tag{6.4}$$

with $x=e^{J/kT}=t^{-1}$, $v=x^{\sec\omega}$ as in reference 25.

An equivalent cluster is built from the z pairs involving one particular spin. As in (4.7) the probabilities for this central atom are, using (6.3)

$$p^R:p^W=\nu^z(1+X_+)^z:(1+X_-)^z. \tag{6.5}$$

Here, ν is again related to the energy difference between right and wrong spins arising from the ordering field. The consistency equation obtained from the B-P method gives

$$\nu^z(1+X_+)^{z-1}=(1+X_-)^{z-1}, \tag{6.6}$$

and comparing this with the equation (57) of reference 25 obtained by the constant coupling approximation

$$\nu=\lambda=e^{-2\beta H_1/kT}=x^{-\tan\omega}. \tag{6.7}$$

It is related to W in that reference by $\lambda^z=W^{1-z}$.

The difference equation for $P^{RR}(0,\mathbf{R})$, etc., can now be obtained as in Sec. 3 by varying the effective field on the pairs around $\mathbf{R}$ by a small space dependent component in the z direction, and comparing the results for an atom on the outside of a cluster with one in the center. With the superlattice structure present, $\gamma'^{zz}(\mathbf{R})$ is defined as in (5.7) and given by the difference equation (5.8). After manipulation we find

$$a^2\kappa_{1z}^2=\frac{(2+X_++X_-)}{(1+X_+)(1+X_-)}\left[\frac{C}{D}-z\right] \tag{6.8}$$

where

$$C/D=\frac{\partial}{\partial H_1}\ln\frac{1+X_+}{1+X_-}\Big/\frac{\partial}{\partial H_1}\ln\frac{\lambda(1+X_+)}{(1+X_-)}$$

$$=\left[1-\frac{\ln x\left[1+\frac{1}{2}x\left(v+\dfrac{1}{v}\right)-\frac{1}{2}x\left(v-\dfrac{1}{v}\right)\sin\omega\right]}{\left[2x^2\ln x\sin^2\omega+x\left(v-\dfrac{1}{v}\right)\cos^3\omega\right]}\right]^{-1}$$

and

$$p_0(z)=(a^2/4r_{1z}^2)=C/4D(2+X_++X_-). \tag{6.9}$$

---

[24] Y. Y. Li, Phys. Rev. 84, 721 (1951).

[25] P. W. Kasteleijn and J. Van Kranendonk, Physica 22, 367 (1956).

In the evaluation of $p_0(z)$ using (3.36) χ is not the magnetic susceptibility because of the staggered spin arrangement.

The x component can also be obtained by the method used in Sec. 4. A small effective field $h_\beta{}^x(\mathbf{R})$ in the x direction is now considered to act on the cluster. To first order the energies and eigenstates referred to the axis of the resultant field at 0 to x, are given by (6.2). Transforming to states referred to the x axis as given by (4.15)

$$
\begin{aligned}
p^{RR}:p^{WW}:p^{RW}:p^{WR}={}&1+\tfrac{1}{2}x\left(v+\frac{1}{v}\right)+\tfrac{1}{2}x\left(v-\frac{1}{v}\right)\cos\omega+x\left(v-\frac{1}{v}\right)\sin\omega\,\sin\theta:1+\tfrac{1}{2}x\left(v+\frac{1}{v}\right)\\
&+\tfrac{1}{2}x\left(v-\frac{1}{v}\right)\cos\omega-x\left(v-\frac{1}{v}\right)\sin\omega\,\sin\theta:1+\tfrac{1}{2}x\left(v+\frac{1}{v}\right)-\tfrac{1}{2}x\left(v-\frac{1}{v}\right)\cos\omega:1+\tfrac{1}{2}x\left(v+\frac{1}{v}\right)-\tfrac{1}{2}x\left(v-\frac{1}{v}\right)\cos\omega
\end{aligned}
\tag{6.10}
$$

to first order in $\sin\omega\,\sin\theta$ which is always small. Comparing this with (6.3) when $\lambda=1+\epsilon$ and $\omega=0$, the expressions required may be written down from (6.8) and (6.9). They are

$$
a^2\kappa_{1x}{}^2=\frac{1+\frac{1}{2}x\left(v+\frac{1}{v}\right)-\frac{1}{2}x\cos\omega\left(v-\frac{1}{v}\right)}{1+\frac{1}{2}x\left(v+\frac{1}{v}\right)}\left[\frac{x\left(v-\frac{1}{v}\right)\cos\omega}{x\left(v-\frac{1}{v}\right)\cos\omega-\ln x\left[1+\frac{1}{2}x\left(v+\frac{1}{v}\right)\right]}-z\right],
\tag{6.11}
$$

and

$$
p_0(x)=\frac{a^2}{4r_{1x}{}^2}=\frac{2x\left(v-\frac{1}{v}\right)\cos\omega\left[1+\frac{1}{2}x\left(v+\frac{1}{v}\right)-\frac{1}{2}x\left(v-\frac{1}{v}\right)\cos\omega\right]}{\left[1+\frac{1}{2}x\left(v+\frac{1}{v}\right)\right]\left[x\left(v-\frac{1}{v}\right)\cos\omega-\left\{1+\frac{1}{2}x\left(v+\frac{1}{v}\right)\right\}\ln x\right]}
\tag{6.12}
$$

Above T_c there is no anisotropy and

$$
a^2\kappa_1{}^2=\frac{4}{x^2+3}\left[\frac{x^2-1}{(x^2-1)-\frac{1}{2}(x^2+3)\ln x}-z\right],
\tag{6.13}
$$

$$
p_0=\frac{a^2}{4r_1{}^2}=\frac{(x^2-1)}{(x^2+3)[(x^2-1)-\frac{1}{2}(x^2+3)\ln x]}.
\tag{6.14}
$$

Putting these expressions into the scattering cross section (1.2) we obtain a result analogous to (4.21) except that because of the superlattice structure we use (5.9) instead of (3.39). The critical scattering in antiferromagnetic materials is therefore peaked around the magnetic superlattice reflections and not about the Bragg reflections nor the forward direction. The expressions given above show that this scattering is of the same order of magnitude and general behavior with absolute temperature as is given by the constants for the ferromagnetic case plotted in Figs. 2 and 3 for the b.c.c. lattice.

7. ISING FERROMAGNET $S=1$

In the preceding sections the critical scattering has been examined for the most important classes of substance where it may be observed, but for simplicity attention was confined to spin $\frac{1}{2}$. Many magnetic substances which are possible for experimental observation have larger spin values and it seemed important to extend the theory at least as far as $S=1$ to obtain some indication of the effects at larger S and in particular to make comparisons with the experiments on iron. As has been shown by earlier calculations the theory is considerably more complicated.[19,20] These complications are shown up by the Ising model with $S=1$ which is now considered in detail.

In this problem we confine attention to the z components of spin which, on each atom, has three possible values 1, 0, -1. It is necessary to introduce two ordering parameters λ^2, ϕ in order to build a consistent theory. One of these can be regarded as arising in a pair or cluster from an effective magnetic field H_1, but this essentially only affects the ordering by altering the relative amounts of 1 and -1 spins. The second reflects a different kind of order by affecting the number of 0 spins. An effective Hamiltonian for a pair may be written

$$
\begin{aligned}
-2JS^z(\mathbf{R})S^z(\mathbf{R}+\boldsymbol{\rho})-2\beta H_1(S^z(\mathbf{R})+S^z(\mathbf{R}+\boldsymbol{\rho}))&\\
+D(S^{z^2}(\mathbf{R})+S^{z^2}(\mathbf{R}+\boldsymbol{\rho})-2).&
\end{aligned}
\tag{7.1}
$$

For a cluster calculation this is summed over all $\boldsymbol{\rho}$ and the H_1 and D terms in $S^z(\mathbf{R})$ are omitted as in (3.3). Proceeding in the B-P fashion, the relative probabilities of finding 1, 0, and -1 at the center of a cluster are

$$
\begin{aligned}
Z^1&=(1+t\phi+t^2\lambda^2)^z,\\
Z^0&=t^z(1+\phi+\lambda^2)^z,\\
Z^{-1}&=(t^2+t\phi+\lambda^2)^z,
\end{aligned}
\tag{7.2}
$$

where in terms of (7.1)

$$t=\exp(-2J/kT),\quad \lambda=\exp(-2\beta H_1/kT),$$
$$\phi=\lambda\exp(-D/kT).$$

By the equivalence of spins at the center and the periphery of a cluster one obtains three consistency equations, of which two are independent,

$$\begin{aligned}Z^1&=\frac{Z^1}{(1+t\phi+t^2\lambda^2)}+\frac{Z^0}{(1+\phi+\lambda^2)}+\frac{Z^{-1}t^2}{(t^2+t\phi+\lambda^2)},\\ Z^0&=\frac{Z^1t\phi}{(1+t\phi+t^2\lambda^2)}+\frac{Z^0\phi}{(1+\phi+\lambda^2)}+\frac{Z^{-1}t\phi}{(t^2+t\phi+\lambda^2)}.\end{aligned}\tag{7.3}$$

These reduce to

$$\begin{aligned}\lambda^2t^{z-1}(1+\phi+\lambda^2)^{z-1}&=\phi\lambda^2(1+t\phi+t^2\lambda^2)^{z-1}\\ &=\phi(t^2+t\phi+\lambda^2)^{z-1}.\end{aligned}\tag{7.4}$$

This equation can also be obtained from the pair Hamiltonian by minimizing the free energy with respect to H_1 and D, i.e., by using the Guggenheim method.[20,21]

One solution of (7.4) is always

$$\lambda=1;\quad t^{z-1}(2+\phi)^{z-1}=\phi(1+t\phi+t^2)^{z-1}.\tag{7.5}$$

Even for this disordered solution ϕ is not unity. Above a critical temperature when $t=t_c$ this is the only solution of (7.4) but below T_c there is a further solution $\lambda<1$ describing the ordered state. Thus, t_c is given by the intersection of curve (7.5) with

$$t\phi=z(1-t^2)-2.$$

The Curie temperature is found to be $kT_c=4.79J$.

To calculate γ' we consider the clusters including $\mathbf{R}$ with ordering parameters modified by the fixed orientation of the spin at 0 in state $i=1, 0, -1$

$$\begin{aligned}\lambda^2&\to\lambda^2+\epsilon_\alpha{}^i(\mathbf{R}'),\\ \phi&=\phi+\delta_\alpha{}^i(\mathbf{R}'),\end{aligned}\tag{7.6}$$

where α labels the neighbors of $\mathbf{R}'$. Then

$$P^{i,j}(0,\mathbf{R})=\frac{Z^iY^{ij}(\mathbf{R})}{\sum_i Z^iY^{ij}(\mathbf{R})},\tag{7.7}$$

where

$$\begin{aligned}Y^{i1}(\mathbf{R}')&=\textstyle\prod_\alpha\left[1+t\phi+t^2\lambda^2+t\delta_\alpha{}^i(\mathbf{R}')+t^2\epsilon_\alpha{}^i(\mathbf{R}')\right],\\ Y^{i0}(\mathbf{R}')&=\textstyle\prod_\alpha\left[1+\phi+\lambda^2+\delta_\alpha{}^i(\mathbf{R}')+\epsilon_\alpha{}^i(\mathbf{R}')\right]t^z,\\ Y^{i-1}(\mathbf{R}')&=\textstyle\prod_\alpha\left[t^2+t\phi+\lambda^2+t\delta_\alpha{}^i(\mathbf{R}')+\epsilon_\alpha{}^i(\mathbf{R}')\right].\end{aligned}\tag{7.8}$$

To first order in ϵ and δ the probabilities (7.7) may be expressed in terms of the sums

$$\sigma^i(\mathbf{R}')=\textstyle\sum_\alpha \epsilon_\alpha{}^i(\mathbf{R}');\quad \zeta\,(\mathbf{R}')=\sum_\alpha \delta_\alpha\,(\mathbf{R}').\tag{7.9}$$

From consistency equations like (3.17)

$$P^i(0)=P^i(\mathbf{R})=P^i(\mathbf{R}+\boldsymbol{\beta})\tag{7.10}$$

for all i, $\boldsymbol{\beta}$, it can be shown that

$$\textstyle\sum_i Z^i\delta_\beta{}^i(\mathbf{R})=\sum_i Z^i\epsilon_\beta{}^i(\mathbf{R}').\tag{7.11}$$

As in Sec. 3, $P^{ij}(0,\mathbf{R}+\boldsymbol{\beta})$ are calculated in two ways and simultaneous difference equations are obtained in the form

$$\begin{aligned}b^jZ^j\textstyle\sum_\beta \zeta^i(\mathbf{R}+\boldsymbol{\beta})+c^jZ^j\sum_\beta \sigma^i(\mathbf{R}+\boldsymbol{\beta})&\\ =B^j\zeta^i(\mathbf{R})+C^j\sigma^i(\mathbf{R})&\end{aligned}\tag{7.12}$$

of which two are independent, say, $j=1$, 0. b^j, c^j, B^j, C^j are complicated coefficients which are determined numerically. At large distances (7.12) may be replaced by differential equations with solutions

$$\begin{aligned}\sigma^i(\mathbf{R})&=h^ie^{-\kappa_1R}/R+j^ie^{-\kappa_2R}/R,\\ \zeta^i(\mathbf{R})&=g^ie^{-\kappa_1R}/R+k^ie^{-\kappa_2R}/R,\end{aligned}\tag{7.13}$$

where h^i, j^i, g^i, k^i are constant coefficients and κ_1, κ_2 the roots of

$$\begin{vmatrix}B^1-b^1Z^1(z+a^2\kappa^2) & C^1-c^1Z^1(z+a^2\kappa^2)\\ B^0-b^0Z^0(z+a^2\kappa^2) & C^0-c^0Z^0(z+a^2\kappa^2)\end{vmatrix}=0.\tag{7.14}$$

When this equation is evaluated numerically it is found that one root $a^2\kappa_1{}^2$ goes to zero at the Curie temperature but the other $a^2\kappa_2{}^2$ is always large (>200) so that the corresponding parts of the solutions (7.13) may be neglected. Equations (7.11) and (7.12) together with the symmetry condition, $P^{ij}(0,\mathbf{R})=P^{ji}(0,\mathbf{R})$ determine the coefficients in (7.13) except for a scaling factor. This can be determined by relating $\sum_{(\mathbf{R})}\langle S_0{}^zS^z(\mathbf{R})\rangle$ to the susceptibility as in (3.36). This susceptibility has been determined by a straightforward but tedious calculation using the B-P method. Writing the final result

$$\langle S_0{}^zS^z(\mathbf{R})\rangle=\langle S_0{}^z\rangle^2+\frac{V_0}{4\pi r_1{}^2}\frac{e^{-\kappa_1R}}{R}\tag{7.15}$$

the parameters κ_1, r_1 are plotted in Figs. 4 and 5.

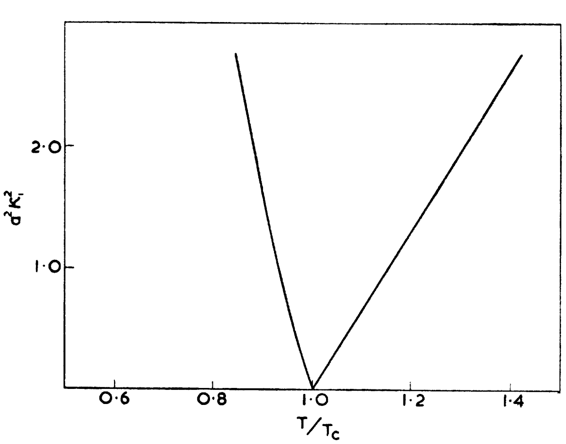

FIG. 4. $a^2\kappa_1{}^2$ for the Ising model, spin 1.

8. HEISENBERG FERROMAGNET $S=1$

This model is probably a good approximation for iron on which there have been a number of experimental observations, which can be compared with the theory. Section 7 has demonstrated the complexity of calculations for $S=1$ but fortunately above T_c a considerable simplification is possible. Below T_c however the calculations were more difficult and the results have only been obtained with further approximations.

The effective Hamiltonian for a pair may be written by analogy with (7.1) as

$$-2J\mathbf{S}(\mathbf{R})\cdot\mathbf{S}(\mathbf{R}+\boldsymbol{\beta})-2\beta H_1\cdot[\mathbf{S}^z(\mathbf{R})+\mathbf{S}^z(\mathbf{R}+\boldsymbol{\beta})] +D[\mathbf{S}^{z^2}(\mathbf{R}+\boldsymbol{\beta})+\mathbf{S}^{z^2}(\mathbf{R})-2]. \quad (8.1)$$

Neglecting D for the moment the eigenstates for the pair can be labeled by the total spin $S=2$, 1, 0 and component S^z. The energy of $|S,S^z\rangle$ is

$$-2J[S(S+1)-2]-2\beta H_1 S^z. \quad (8.2)$$

Expanding the states in the representation $|S^z(\mathbf{R}), S^z(\mathbf{R}+\boldsymbol{\beta})\rangle$ the relative probabilities P^{ij} that these neighbors will be in the various spin states are found.

$$P^{11}:P^{10}:P^{1-1}:P^{-10}:P^{-1-1}:P^{00} =1/\lambda^2:\tfrac{1}{2}(1+t^2)/\lambda:(1+3t^2+2t^3)/6 :\tfrac{1}{2}(1+t^2)\lambda:\lambda^2:(2+t^3)/3, \quad (8.3)$$

where λ and t are defined in (7.2).

As in Sec. 4 it is now possible to build an equivalent cluster where with the definition (7.2)

$$Z^1=\left[\frac{1}{\lambda}+\tfrac{1}{2}(1+t^2)+\tfrac{1}{6}\lambda(1+3t^2+2t^3)\right]^z,$$

$$Z^0=\left[\tfrac{1}{2}(1+t^2)\left(\frac{1}{\lambda}+\lambda\right)+\tfrac{1}{3}(2+t^3)\right]^z, \quad (8.4)$$

$$Z^{-1}=\left[\frac{1}{6\lambda}(1+3t^2+2t^3)-\tfrac{1}{2}(1+t^2)+\lambda\right]^z.$$

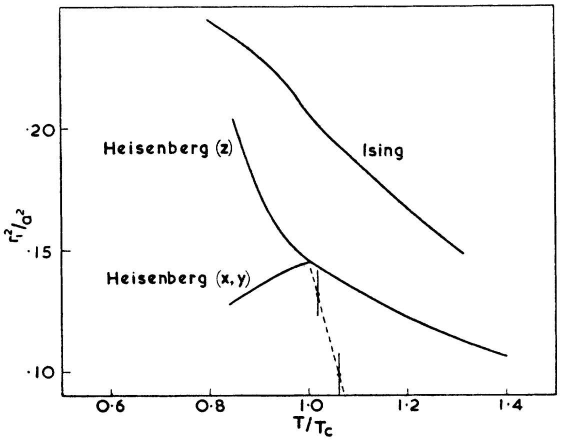

FIG. 5. r_1^2/a^2 for the Ising and Heisenberg models, spin 1. The Heisenberg curves for $T<T_c$ have been calculated on an approximate theory and may not be reliable.

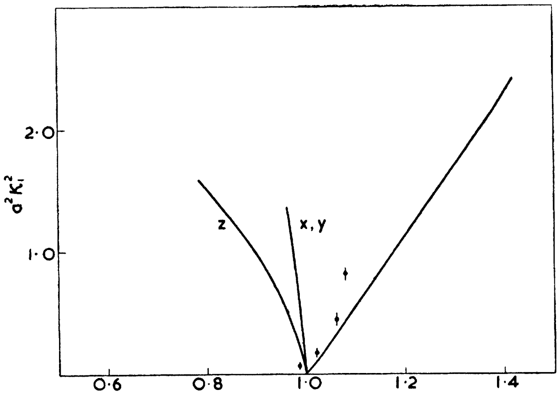

FIG. 6. $a^2\kappa_1^2$ for the Heisenberg model, spin 1. The curves for $T<T_c$ have been calculated on an approximate theory and may not be reliable.

The consistency equations may be determined by either a B-P method or the constant coupling method and are

$$\lambda[\tfrac{1}{2}(1+t^2)(1+\lambda^2)+\lambda\tfrac{1}{3}(2+t^3)]^{z-1} =\lambda^2[1+\tfrac{1}{2}\lambda(1+t^2)+\tfrac{1}{6}\lambda^2(1+3t^2+2t^3)]^{z-1} =[\tfrac{1}{6}(1+3t^2+2t^3)+\tfrac{1}{2}\lambda(1+t^2)+\lambda^2]^{z-1}, \quad (8.5)$$

which clearly cannot in general be satisfied. They are satisfied however by $\lambda=1$ so that at high temperatures $H_1=0$, $D=0$, i.e., no ordering, is a solution. Taking any pair of equations they also determine a unique transition temperature below which another root would occur, *viz.*,

$$z(5-3t_c^2-2t_c^3)=3(5+t_c^2),$$

where $t_c=\exp(-2J/kT_c)$.

Below this temperature a consistent theory demands that both H_1 and D be nonzero, but close to T_c when H_1 is small the equations are satisfied to first order so D must be of order $(\beta H_1)^2/kT_c$. Because of the noncommutation of $\mathbf{S}(\mathbf{R})$ the problem including the two parameters is much more tedious than is Sec. 7 and has not been pursued. Above T_c, however, it is possible to use the one parameter theory. As in Sec. 3, one calculates $P^{ij}(0,\mathbf{R})$ by considering that the fixing of the spin at 0 induces an ordering field on the clusters around $\mathbf{R}$ and λ becomes $1+\epsilon_\beta(\mathbf{R})$. There is also a second ordering effect through D but it is an order of magnitude smaller than that through H_1 and is neglected. Following through the argument of the earlier sections we find

$$a^2\kappa_1^2=\frac{(5+9t^2+4t^3)}{2(5+3t^2+t^3)}\left[\frac{3(5+t^2)}{(5-3t^2-2t^3)}-z\right] \quad (8.6)$$

$$p_0=\frac{(5+9t^2+4t^3)(5+t^2)}{(5+3t^2+t^3)(5-3t^2-2t^3)}=\frac{2a^2}{3r_1^2} \quad (8.7)$$

if r_1 is defined as in reference 10. The cross section is given by (4.21) with these substitutions, and is independent of **e**. Comparing the plots for the b.c.c. lattice in Figs. 5 and 6 with those in Figs. 2 and 3 giving the results for $S=\frac{1}{2}$; κ_1 and r_1 do not differ greatly with S value. In particular at T_c itself r_1 has the same value as given by (3.45).

Just below T_c, a one parameter theory can be constructed by applying a more approximate theory which may not be very reliable, similar to that developed by Weiss.[19] D is again taken to be zero and all the ordering is produced by the effective magnetic field H_1. It is not now possible to satisfy the condition that a central and a peripheral spin have the same probabilities of being in each spin state since this leads to two equations in one unknown (8.2). Instead, a single consistency condition is imposed that the average magnetic moment on the two spins be the same; i.e.,

$$Z^1-Z^{-1}=\frac{Z^1(1-b\lambda^2)}{1+a\lambda+b\lambda^2}+\frac{Z^0(1-\lambda^2)a}{a(1+\lambda^2)+c\lambda}+\frac{Z^{-1}(b-\lambda^2)}{b+a\lambda+\lambda^2}, \quad (8.8)$$

where

$$a=\tfrac{1}{2}(1+t^2), \quad b=\tfrac{1}{6}(1+3t^2+2t^3), \quad c=\tfrac{1}{3}(2+t^3). \quad (8.9)$$

The $\gamma'(\mathbf{R})$ are calculated by the same method as in the earlier sections except that it is essential to calculate the change in magnetic moment induced in a spin at R by fixing spin 0 in state i, i.e., $P_{0,R}{}^{i1}-P^{i-1}(0,\mathbf{R})$; rather than $P^{ij}(0,\mathbf{R})$ separately. For $\gamma'^{zz}(\mathbf{R})$ the effective fields on the pairs participating in the cluster around $\mathbf{R}$ have an additional component $h_\beta{}^z(\mathbf{R})$ and λ becomes $\lambda+\epsilon_\beta(\mathbf{R})$. The usual difference equation (3.23) is reproduced. The calculation is straightforward but tedious and gives finally

$$a^2\kappa_{1z}{}^2=\{Z^1\alpha^1+Z^0\alpha^0+Z^{-1}\alpha^{-1}\}\left\{\frac{\lambda Z^1(\lambda^2-b)}{(1+a\lambda+b\lambda^2)}\right.$$
$$\left.-\frac{\lambda Z^{-1}(b\lambda^2-1)}{(b+a\lambda+\lambda^2)}+\frac{\lambda^2(Z^1-Z^{-1})^2}{Z^1+Z^0+Z^{-1}}\right\}^{-1} \quad (8.10)$$

$$p_0(z)=\frac{2a^2}{3r_{1z}{}^2}=z\left\{\frac{Z^1(2b+a\lambda)}{(1+a\lambda+b\lambda^2)}\right.$$
$$\left.+\frac{Z^{-1}(2b\lambda^2+a\lambda)}{(b+a\lambda+\lambda^2)}\right\}(Z^1+Z^0+Z^{-1})^{-1}$$
$$+a^2\kappa_{1z}{}^2[(Z^1+Z^{-1})Z^0+4Z^1Z^{-1}]$$
$$\times[Z^1+Z^0+Z^{-1}]^{-2}, \quad (8.11)$$

where

$$\alpha_1=z\lambda(2b+a\lambda)(b-\lambda^2)+\lambda^2(a\lambda^2+4b\lambda+ab)$$
$$\alpha_0=z(1-\lambda^2)^2a^2+a\lambda(c\lambda^2+4a\lambda+c) \quad (8.12)$$
$$\alpha_{-1}=z\lambda^2(2b\lambda+a)(b\lambda^2-1)+\lambda^2(ab\lambda^2+4b\lambda+a).$$

A few values have been calculated and extrapolated curves are given in Figs. 5 and 6 for the b.c.c. lattice.

For $\gamma'^{xx}(\mathbf{R})$ the calculation is similar to that given for this quantity in Sec. 4. The results have the form (8.10) with $\lambda=1$ and (8.9) replaced by

$$a=2[\lambda^2-\lambda+1+\lambda x^2]/(1+\lambda)^2$$
$$b=\left(\lambda^2+\frac{1}{\lambda^2}+\frac{2}{3}+8x^2+\frac{16}{3}x^3\right)\lambda^2/(1+\lambda)^4 \quad (8.13)$$
$$c=4\left(\lambda^2+\frac{1}{\lambda^2}+\frac{2}{3}+\frac{4}{3}x^3\right)\lambda^2/(1+\lambda)^4$$

and are plotted in Figs. 5 and 6. The cross section is given by (4.21) with these substitutions.

This approximation is only valid in the region just below T_c where D is small, and is possibly adequate if $|T_c-T|<0.1T_c$. In this region the curves of κ_1 and r_1 again resemble the results in Figs. 2 and 3 for $S=\frac{1}{2}$. The experimental points determined by Shull, Wilkinson, and Gersch[3,4] are shown in the figures. The agreement is good near T_c but at higher temperatures the agreement is less satisfactory. The discrepancy may possibly be accounted for by the inelasticity of the scattering which will be discussed in a subsequent paper. Lowde's[5] results also confirm the general form and order of magnitude of the predicted scattering around a Bragg reflection. Shull *et al.* find no dependence of the cross section on e_z below T_c but have made measurements only at one angle. Lowde has some indication of a small change with magnetization of the kind predicted. Further experimental observation of this effect would be most interesting.

9. CONCLUSION

A method has been given for determining the correlation functions between spin components at all distances $\mathbf{S}^i(0)\mathbf{S}^j(\mathbf{R})$ in ordered magnetic materials. Details of these functions are given for a number of the simple cases—ferromagnets with spin $\frac{1}{2}$ and 1, antiferromagnets with spin $\frac{1}{2}$, and binary alloys. The parameters κ_1 and r_1 describing the order are found to differ by only a factor of about two and the above results are probably a useful first approximation for other spin values which may be observed experimentally. The method could be readily extended in some directions, for example to magnets of arbitrary spin above T_c.

The correlations have been used to calculate the critical scattering of neutrons close to T_c. There is reasonable agreement with the experimental results on iron and the Heisenberg model with $S=1$ and it would be interesting to have experimental results on the predicted dependence on the direction of the magnetization below T_c, and on the predicted effects on antiferromagnets and binary alloys. The inelastic scattering arising from the time dependence of the correlation will be discussed in a later paper.

The correlation functions have an important bearing on other properties, and in particular the modification of the specific heat anomaly and the effect on the electrical resistivity of magnets are being examined.

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Theory of the Magnetic Scattering of Neutrons by CoO

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1. INTRODUCTION

THE spin arrangement in the antiferromagnetic state of cubic crystals MnO, FeO, CoO, and NiO has been a subject of active interest during the past several years. The first neutron diffraction study by Shull, Strauser, and Wollan[1] indicated that the spin superstructure in these oxides is such that a sheet of plus spins and a sheet of minus spins alternate along one of the [111] directions (a structure suggested by Néel[2]) and that the direction of the spins is one of the [100] axes, except in FeO in which it was found to be perpendicular to those sheets, i.e., parallel to [111]. The electronic state of a free Mn^{++} is $(5d)^5\,{}^6S$ and so one expects the magnetic anisotropy energy in MnO to arise principally from the magnetic dipole-dipole interactions among the manganous ions, as the electronic state ${}^6S$ would suffer little change in the crystal. A simple elementary consideration, as well as an exact calculation due to Kaplan,[3] of these interactions shows that the stable spin direction should lie in the (111) plane, in contradiction to the experiment. A similar situation is encountered with NiO, since the orbital moment of Ni^{++}, whose electronic state is $(3d)^8\,{}^3F$, is to be quenched in the cubic crystalline electric field of the oxide. A new neutron diffraction study carried out by Roth[4] has revealed features which are in favor of the theory. His results indicate that the spin direction in MnO is either $[\bar{1}11]$ or $[\bar{1}10]$, the latter in the (111) plane, and that in NiO $[\bar{1}10]$. Both experiments, new and earlier, conclude that the spin direction in FeO is perpendicular to the (111) sheets, as reflections from these planes vanish. This could be expected from a theory cited below. In CoO, Roth's preliminary interpretation of his results was that the spin direction might be either $[11\bar{2}]$ or $[0\bar{1}1]$, both being in the (111) plane, which, however, contradicts the theory. The situations with FeO and CoO are somewhat complicated due to the existence of a residual orbital moment of the ions in the crystal. This was already conceived by Shull *et al.*[1] in connection with larger observed intensities of diffraction lines than expected on the basis of the spin only theory.

The electronic states of a free Fe^{++} and a free Co^{++} are, respectively, $(3d)^6\,{}^5D$ and $(3d)^7\,{}^4F$. In the cubic crystalline field of the oxides, their orbital degeneracy is lifted only partially leaving the ground-state triply degenerate in both cases. This ground state is described in terms of an angular momentum operator $\mathbf{l}$ of magnitude one, i.e., the total angular momentum operator $\mathbf{L}$ of the free ion can be replaced by $-\mathbf{l}$ in FeO and $-\frac{3}{2}\mathbf{l}$ in CoO. This small $\mathbf{l}$ is coupled with the spin angular momentum, $\mathbf{S}$, to form a total angular momentum $\mathbf{l}+\mathbf{S}$, and the eigenstates of the ions are characterized by the eigenvalues of $(\mathbf{l}+\mathbf{S})^2=j(j+1)$, with $j=1, 2, 3$ in the case of FeO and $j=\frac{1}{2}, \frac{3}{2}, \frac{5}{2}$ in the case of CoO, if no exchange couplings exist among the ions. Actually they do exist, and when taken into account in the approximation of the Weiss molecular field, states with different j but with the same l_z+S_z couple where z is the direction of the molecular field. Kanamori[5] has shown that the state of Co^{++} in CoO at absolute zero is described by the wave function

$$\psi=a\psi_{\frac{3}{2},-1}+b\psi_{\frac{1}{2},0}+c\psi_{-\frac{1}{2},1} \tag{1}$$

with

$$a=0.900,\quad b=-0.401,\quad c=0.169, \tag{2}$$

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