data obtained in the region above the Curie point (122°C). Similar data were obtained for (80%Ba-20%Sr)TiO_3.

The dispersion is best displayed by showing the permittivity as a function of frequency. Figure 2 shows the dispersion observed using polycrystalline (Ba-Sr) TiO_3 samples (Curie temperature 55°C). Note that the dispersion is not a simple relaxation process, but exhibits resonance characteristics as the Curie point is approached from the ferroelectric side. No dispersion is apparent above the Curie point; one curve in this region is also shown in Fig. 2. The data displayed in Fig. 2 were obtained using two samples of incommensurate lengths of the same material. The appearance of resonance character in the permittivity spectrum recalls Kittel's theory of domain boundary inertia.[5] It appears that the domain walls respond in a manner similar to a forced damped harmonic oscillator.

Further measurements are in progress to determine the effect of biasing electric fields on the dispersion of (Ba-Sr)TiO_3 ferroelectrics. The effect of grain size on the spectrum is also of interest to examine the possibility of acoustical resonance.

[5] C. Kittel, Phys. Rev. **83**, 458(1951).

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Antiferromagnetic Susceptibility of the Plane Triangular Ising Lattice

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The magnetic moment transformation developed by Fisher enables the antiferromagnetic susceptibility of the plane triangular Ising lattice to be expanded as a power series that converges over the whole temperature range $0 \leqslant T \leqslant \infty$. The dominant asymptotic behavior of the coefficients conjectured from extrapolations by Domb and Sykes, and independently by Park, has been established theoretically by Fisher. A counting theorem based on the method of Oguchi enables the first twelve terms of the expansion to be derived. It is found possible to evaluate the susceptibility numerically over the whole temperature range with a maximum error of 0.1% at $T=0$. It is concluded that the specific susceptibility per spin $(kT\chi_0/m^2)$ falls smoothly from unity at $T=\infty$ to a value at $T=0$ which does not differ by more than 0.1% from 5/36, and the form of the counting theorem leads it to be surmised that it is exactly 5/36.

1. INTRODUCTION

No exact closed analytical expression has yet been given for the zero-field susceptibility of any two-dimensional Ising model. It is the purpose of the present paper to evaluate the antiferromagnetic susceptibility of the plane triangular lattice numerically from exact series developments.

At high temperatures the reduced zero-field susceptibility χ defined as $kT\chi_0/m^2$ may be expanded in powers of the high-temperature counting variable $v=\tanh(J/kT)$, by the method of Oguchi[1,2] as

$$\chi(v)=\sum_n a_n v^n, \quad a_0=1. \tag{1}$$

The first 12 coefficients have been obtained by Sykes.[2] The a_n are positive integers and the series converges[3] only for $|v|\leqslant v_f$, where $v_f=\tanh(J/kT_f)=0.267949$ and T_f is the ferromagnetic Curie temperature. The series (1) can only be used to estimate $\chi(v)$ in the range $|v|\leqslant v_f$, i.e., $T_f\leqslant T\leqslant\infty$.

At $T=0$ which corresponds to $v=-1$ the ground state of the antiferromagnetic triangular lattice is highly degenerate[4] and no series or asymptotic developments about this origin have so far been given.

Recently Fisher[5] has developed a magnetic moment transformation that relates the reduced susceptibility of the triangular lattice to that of the honeycomb lattice. If χ_T denotes the reduced susceptibility of the triangular lattice and χ_H that of the honeycomb lattice, then

$$\chi_T(v)=\tfrac{1}{2}[\chi_H(w)+\chi_H(-w)], \tag{2}$$

for

$$w^2=v(1+v)/(1+v^3). \tag{3}$$

Equation (2) relates the susceptibility of the triangular lattice at temperature v to the mean of the ferromagnetic and antiferromagnetic susceptibilities of the honeycomb lattice at a temperature w determined by (3). As v varies from 0 to -1, w^2 varies from 0 to $-\frac{1}{3}$ and the high-temperature honeycomb susceptibility series corre-

[1] T. Oguchi, J. Phys. Soc. Japan **6**, 31 (1951).
[2] M. F. Sykes, J. Math. Phys. **2**, 52 (1961).
[3] C. Domb and M. F. Sykes, J. Math. Phys. **2**, 63 (1961).
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sponding to (1),

$$\chi_H(w)=\sum_n b_n w^n, \quad b_n=1, \tag{4}$$

converges for $-\frac{1}{3}\leqslant w^2\leqslant 0$ since for the honeycomb lattice $v_f=1/\sqrt{3}$. Thus the series development of the right-hand side of (2), which we shall denote by $\chi_T(w)$, will converge over the whole antiferromagnetic temperature range $0\geqslant v\geqslant -1$. The expansion of $\chi_T(w)$ has been derived up to the term in w^{24} by Sykes[2] using a special counting theorem. The asymptotic behavior of the coefficients is accurately known[6] and it is therefore possible to evaluate the function $\chi(w)$ numerically with high accuracy over the whole range; this is undertaken in the next section.

Similar transformations can be applied to the antiferromagnetic Kagomé lattice[5,7] and we shall evaluate the corresponding reduced susceptibility $\chi_K(v)$.

We shall compare the results obtained with the behavior of some standard approximations and other numerical extrapolation techniques.

2. SUMMATION OF THE SERIES $\chi_T(w)$

The series to be summed is[2]

$$\chi_T(w)=\sum_n c_n w^{2n}=1+6w^2+24w^4+90w^6+318w^8 +1098w^{10}+3696w^{12}+12\,270w^{14} +40\,224w^{16}+130\,650w^{18}+421\,176w^{20} +1\,348\,998w^{22}+4\,299\,018w^{24}+\cdots. \tag{5}$$

The asymptotic behavior of the coefficients in high-temperature susceptibility expansions of two-dimensional lattices has been studied by Domb and Sykes[3,8] and by Park[9] who concluded that for large n the a_n of (1) behaved as $n^{3/4}v_f^{-n}$ and this corresponds to a singularity at v_f of the form $(v_f-v)^{-7/4}$. This conclusion has recently received rigorous support from the work of Fisher[6] on the simple quadratic lattice and we shall suppose that these conclusions are exact for the triangular and honeycomb lattices—that is, we shall suppose that in (5)

$$c_n\sim n^{3/4}3^n. \tag{6}$$

The extent to which the coefficients conform to this limiting behavior for the values of n at our disposal may be judged from an examination of the quantity $\theta_n=c_n/c_{n-1}(1+3/4n)$ as proposed by Domb and Sykes[3] which, if (6) is correct, will approach 3 as n increases. The last six values of θ_n are given in Table I and it will be seen that even for small values of n the asymptotic behavior is remarkably well observed. In fact, the series (5) is the most smoothly convergent high-temperature Ising series we have yet encountered.

To evaluate $\chi_T(w)$ we shall employ a device suggested by a paper by Park,[9] which enables the series (5) to be summed accurately. We suppose that near w_c the function $\chi_T(w)$ behaves like $(1-3w^2)^{-7/4}$ and investigate the assumption that we may write

$$\chi_T(w)=\phi(w)(1-3w^2)^{-7/4}. \tag{9}$$

To do this we divide out the second factor in (9) and this is conveniently done logarithmically. We set

$$\ln\phi(w)=\ln\chi_T(w)+(7/4)\ln(1-3w^2), \tag{10}$$

and find

$$\ln\phi(w)=\sum_n d_n w^{2n}=0.75w^2-1.875w^4+2.25w^6 -5.4375w^8+12.15w^{10}-26.625w^{12} +54.1071428w^{14}-109.21875w^{16}+210.75w^{18} -396.375w^{20}+666.0681818w^{22} -923.8125w^{24}. \tag{11}$$

In Table II the successive values of the ratios $r_n=d_n/d_{n-1}$ in (11) are given. It would appear that the quantity r_n is approaching a limit well below 3 and that the assumption that the singularity occurs as *a factor* is correct.

We now write

$$\ln\chi_T(w)=-(7/4)\ln(1-3w^2)+\sum_{r=1}^{12} d_r w^{2r}+R(w), \tag{12}$$

where $R(w)$ denotes the remainder after summation of the first 12 terms of (11). We shall denote the value of $\chi_T(w)$ obtained by neglecting this remainder by $\chi_T(w)_{12}$, and we have

$$\chi_T(w)=\chi_T(w)_{12}\exp R(w). \tag{13}$$

Since we may suppose that the ratios in Table II are monotonic decreasing, we observe that on taking the last ratio and assuming a geometric progression we have

$$1\geqslant \exp R(w)\geqslant 0.9985 \quad \text{for} \quad -1\leqslant v\leqslant 0, \tag{14}$$

and therefore the approximation $\chi_T(w)_{12}$ will be correct to within 15 parts in 10 000. The behavior of the ratios in Table II is not sufficiently regular to suggest any

TABLE I. Values of θ_n for the series $\chi_T(w)$.

n	θ_n
7	2.9985
8	2.9973
9	2.9982
10	2.9988
11	2.9985
12	2.9988

TABLE II. Values of $r_n=d_n/d_{n-1}$ for the expansion of $\ln\phi(w)$.

$r_5=2.2345$	$r_9=1.9296$
$r_6=2.1914$	$r_{10}=1.8808$
$r_7=2.0322$	$r_{11}=1.6804$
$r_8=2.0186$	$r_{12}=1.3870$

[6] M. E. Fisher, Physica **25**, 521 (1959).
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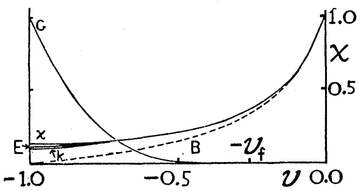

FIG. 1. The reduced antiferromagnetic susceptibility of the plane triangular lattice; (χ) Exact, (*E*) Energetic approximation. (*k*) Kikuchi approximation, (*B*) Bethe approximation, and (*G*) Residual correlation function.

more refined extrapolation. The remainder $R(w)$ will have a maximum value at $T=0$ or $w^2=-\frac{1}{3}$. At this point, from (13) and (14), we calculate

$$0.13884 \leqslant \chi_T(w^2=-\tfrac{1}{3}) \leqslant 0.139093. \quad (15)$$

To complete the extrapolation we observe that a study of the configurational data required to derive the high-temperature series for $\chi_T(v)$ leads to the conclusion[2,10] that we may write

$$\chi(v)=(1-\sigma v)^{-2}[1-(\sigma-1)v+v^2-2vU(v)+G(v)], \quad (16)$$

where $\sigma+1$ is the coordination number, $U(v)$ is the reduced energy $U(0)=0$, $U(1)=q/2$, and $G(v)$ is an unknown function defined in terms of certain lattice configurations but not known in closed form. It is found that the approximation obtained by neglecting $G(v)$, which we shall call the energetic susceptibility (χ_T^E), is correct to within $\frac{1}{2}\%$ in the region $-v_f \leqslant v \leqslant 0$. [In this region the approximation $\chi_T(w)_{12}$ will be correct to within 0.02%.] We must therefore suppose that at high temperatures the energy, which is determined by the first-order correlations between spins, plays a dominant role. At low temperatures, however, the function $G(v)$, which we shall call the residual correlation function, becomes important. By substitution of (15) in (16) we find that at $v=-1$

$$0.9998 \leqslant G(-1) \leqslant 1.0074, \quad (17)$$

and from this we shall conclude $G(-1)$ is very probably unity. We have therefore employed as remainder the expression

$$R(w)=1260w^{26}/(1+1.364w^2), \quad (18)$$

which corresponds to taking a geometric progression after the last term in (11) with a common ratio chosen to make $G(-1)=1$. The relatively small value of the remainder leads us to suppose that the value of $\chi_T(w)$ calculated from (13) and (18) should prove correct to 5 parts in 10 000.

3. ANTIFERROMAGNETIC SUSCEPTIBILITY OF THE PLANE TRIANGULAR LATTICE

By the method of the previous section the susceptibility of the plane triangular lattice has been calculated and we plot the result against the variable v in Fig. 1. On the same figure we also plot $\chi_T^E(v)$ and the residual correlation function $G(v)$. The function $G(v)$ is negative in the region $-0.51372<v<0$, and the difference $\chi_T^E-\chi_T$ has a maximum value in this range of about $\frac{1}{2}\%$ of χ_T near $v=-0.37$ (or $0.7T_f$) and is zero at $v=-0.51372$ ($0.48T_f$) where $G(v)$ changes sign. Thus effectively the approximation $\chi_T^E(v)$ is accurate to within $\frac{1}{2}\%$ in the temperature region $\frac{1}{2}T_f<T<\infty$; at $\frac{1}{4}T_f$ the error is 9% and at $0.1T_f$ it rises to near its maximum value of 20%.

4. ANTIFERROMAGNETIC SUSCEPTIBILITY OF THE KAGOMÉ LATTICE

The relationship between the partition functions of the Kagomé and honeycomb lattices has been studied by Naya[7] and Fisher.[5] The reduced energy $U_K(v)$ and reduced susceptibility $\chi_K(v)$ of the Kagomé lattice can be related to those of the honeycomb lattice. In our notation the results may be written

$$\chi_K(v)=1+v(1-v+v^2)[6\chi_H(w^*) -2U_H(w^*)/3-2]/(1+v^2)^2, \quad (19)$$

$$U_K(v)=2(1+v)^2U_H(w^*)/3(1+v^2) +2v/(1+v^2)-v(1+v)/(1+v^3), \quad (20)$$

for

$$w^*=v(1+v)/(1+v^3). \quad (21)$$

For the antiferromagnetic region $-1<v<0$ the variable w^* varies from 0 to $-\frac{1}{3}$ and is therefore always entirely inside the circle of convergence of the high-temperature expansion of $\chi_H(w^*)$ which is $-1/3^{\frac{1}{2}}<w^*<0$. Since 24 terms[2] of the expansion of $\chi_H(w^*)$ are available, the quantity $\chi_K(v)$ can be evaluated accurately over the whole antiferromagnetic temperature range. By a straightforward numerical extrapolation we estimate

$$\chi_H(-1/3^{\frac{1}{2}})=0.397193\pm0.000002, \quad (22)$$

and for the Kagomé lattice

$$\chi_K(-1)=0.18943\pm0.00001, \quad (23)$$

and

$$G(-1)=0.3643. \quad (24)$$

The quantities $\chi_K(v)$, $\chi_K^E(v)$, and $G(v)$ for the Kagomé lattice are plotted in Fig. 2. The function $G(v)$ is always positive for this lattice.

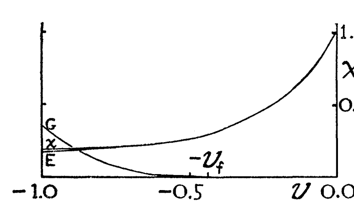

FIG. 2. The reduced antiferromagnetic susceptibility of the Kagomé lattice; (χ) Exact, (*E*) Energetic approximation, and (*G*) Residual correlation function.

[10] M. F. Sykes and M. E. Fisher, Phys. Rev. Letters 1, 321 (1958).

5. APPROXIMATIONS AND EXTRAPOLATIONS

Before examining some of the approximations that have been proposed for the antiferromagnetic susceptibility of the Ising model, we make the following general observation.

The reduced energy of the triangular lattice may be expanded at high temperatures in powers of v as

$$U(v)=3v+6v^2+12v^3+24v^4+54v^5+\cdots \quad (25)$$

This series has a generating function (Houtappel[11])

$$\frac{-(1+v^2)}{2v}\left[\frac{1-3v-3v^2+v^3}{(1-v)^3}\frac{2}{\pi}K(k)-1\right], \quad (26)$$

where $K(k)$ is the complete elliptic integral of the first kind and $k^2=16v^3(1-v+v^2)/(1-v)^6(1+v)^2$. Although the series (25) converges only for $|v|\leqslant v_f$, expression (26) is valid for all $|v|\leqslant 1$. Because of this, the energetic susceptibility can be continued analytically down to $v=-1$ and it seems likely that if the generating function of the series for $\chi_T(v)$ could be recognized it would prove to be the complete solution.

In the standard Bethe approximation the high-temperature expansion for the susceptibility has as generating function the Firgau formula,[12]

$$\chi_B=(1+v)/(1-\sigma v), \quad |v|<1/\sigma. \quad (27)$$

The restriction on the range in (27) results from the assumption in the derivation of χ_B that there is no long-range order. Since no long-range order is possible on the triangular lattice for $J<0$, we take as the Bethe approximation in the low-temperature antiferromagnetic region the analytic continuation of (27) which we plot in Fig. 1. Since this approximation depends only on the coordination number and neglects the detailed structure of the lattice, it is not surprising that it gives only a crude representation at low temperatures.

An approximation which makes some allowance for the structure of the lattice is that proposed by Kikuchi,[13] and the corresponding high-temperature susceptibility has been evaluated by Burley,[14] who finds for the triangular lattice

$$\chi_k(v)=(1-v)/(1-3v)(1-4v), \quad |v|\leqslant\tfrac{1}{4}. \quad (28)$$

Here again the restricted range of v results from the assumption of no long-range order. We plot the function (28) in Fig. 1, and it will be seen that the approximation is almost as good as $\chi_T{}^E$. At $T=0$, (28) gives $\chi_k(-1)=0.100$ while $\chi_T{}^E(-1)=0.111$. We remark in parentheses that the ground state of the triangular lattice is highly degenerate and has a finite entropy[4] of $S/R=0.32306$, while the Kikuchi approximation[13] yields $S/R=0.2877$ which may be regarded as reasonable for so simple an approximation.

At the temperature $T=T_f$, i.e., when $v=-v_f$, we find

$$\begin{aligned}\chi_T(-v_f)&=0.339277,\\ \chi_k(-v_f)&=0.339271,\\ \chi_T{}^E(-v_f)&=0.340157.\end{aligned} \quad (29)$$

The excellent agreement (0.002%) between the first two estimates in (29) is to some extent misleading, since it is fortuitous in much the same way as the agreement between $\chi_T{}^E$ and χ_T at $v=-0.51372$ which results from the change of sign of $G(v)$ at this point.

The deficiencies of the Kikuchi approximation are more evident in the ferromagnetic region where (28) predicts a singularity of type $(v_f-v)^{-1}$ in place of $(v_f-v)^{-7/4}$. Instead of the analytic continuation of an approximate high-temperature formula, we could attempt to continue a representation of the high-temperature series $\chi(v)$. A method of deriving such a representation has been discussed by Park[9] and for details reference should be made to his paper. Essentially it consists in finding a function which generates all the known coefficients correctly. If the true generating function is a simple one, it will be detected and the complete solution obtained in closed form. Park was able to identify the magnetization of the simple quadratic lattice in this way from its series expansion. The method is fraught with danger since an approximation that gives the first n terms of a series correctly is not necessarily a good one. It will be better if it also gives the correct radius of convergence and better still if it has the correct asymptotic behavior. In this respect the function

$$\chi_M(v)=(1-4v+v^2)^{-7/4}(1+3v^2)(1-v)^{\frac{1}{2}}(1+v^2)^{\frac{3}{4}}\times(1+2v^2)^{-2}(1+v)^{-\frac{1}{2}}, \quad (30)$$

which reproduces the first 8 terms of the expansion of $\chi_T(v)$ correctly,[15] may be regarded as an excellent approximation to χ_T for $|v|<v_f$. At $v=-v_f$, (30) gives 0.33917 compared with the exact value of 0.33928; it has the correct radius of convergence (namely the least positive root of the quadratic factor), and this occurs to the correct power. The representation will be satisfactory both for the ferromagnetic and for the antiferromagnetic susceptibility. We shall call such an approximation an "algebraic mimic," and such a mimic provides a convenient method of summation of the series but is quite unsuitable for analytic continuation unless it happens to be the true generating function. In fact Eq. (30) yields the inadmissible value $\chi(-1)=\infty$. Algebraic mimics can be constructed which generate any required number of terms correctly, but we have so far been unable to find any mimic that could be proposed as the exact solution, which as we have already urged could be continued analytically, and it seems un-

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[14] D. M. Burley, Phil. Mag. **5**, 909 (1960).

[15] Ninth term 732 694 in place of 732 678.

likely in view of the form of (26) that $\chi(v)$ would prove to be algebraic.

Another method of mimicking the high-temperature expansion is that employed by Domb and Sykes[8] for the corresponding ferromagnetic problem. They write

$$\chi(v)=A(1-t)^{-7/4}+\psi(t), \quad 1/t=v_c/v, \qquad (31)$$

the polynomial $\chi(t)$ being chosen to reproduce the known terms correctly. The values they quote for the triangular lattice give $\chi(-v_f)=0.3385$ which is correct to 0.23%, but the number of significant figures quoted is inadequate for the antiferromagnetic problem and only 8 terms were at their disposal. With the 12 terms now available, the representation (31) could be improved and the method is again adequate for summation in the high-temperature region. As $T\rightarrow 0$, $t\rightarrow -\infty$ and the form of $\psi(t)$ makes an estimation of $\chi(-1)$ by this method impracticable.

6. CONCLUSIONS

A formula has been given that enables the antiferromagnetic susceptibility of the plane triangular lattice to be evaluated over the entire temperature range with a maximum error of 5 parts in 10 000 at $T=0$. That this has proved possible results from three facts. First, the magnetic moment transformation yields an expansion that converges up to $T=0$. Second, the asymptotic behavior of the coefficients is well established and this makes a reliable summation possible. Third, a counting theorem enables an adequate number of terms of the series to be derived.

We have found that, while adequate for estimating $\chi(v)$ in the range $0>v>-v_f$, none of the approximate methods previously proposed for extrapolating the high-temperature susceptibility series enables the susceptibility to be evaluated at low temperatures. In the range $\frac{1}{2}T_f<T<\infty$, the energetic approximation is the most satisfactory. Unfortunately, although this is probably still true for three-dimensional lattices, the energy is not known exactly in these cases.

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Influence of Silver Impurities on the Annealing Kinetics of Quenched Gold Specimens

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The recovery of quenched-in extra resistivity has been studied in thin gold wires, to which atomic concentrations of silver equal to 1.2×10^{-3} or 1.4×10^{-4} have been added. Recovery occurs at higher temperatures than for pure specimens, the effective activation energy being larger than 1 ev. The interpretation is that vacancy-impurity complexes are formed, whose binding energy is about 0.3 ev. Evidence of motion of defects at low temperature is also obtained in the case of impure specimens.

I. INTRODUCTION

A GREAT deal of attention has been devoted in the last few years to the kinetics of lattice vacancies in face-centered cubic metals. After the first results by Koehler *et al.*[1] for gold, the quenching method has been widely used to inject vacancies into the specimens; the features of recovery of quenched-in extra resistivity during annealing at suitable temperatures have been assumed to be directly related to the behavior of lattice vacancies.

Experimental evidence has been accumulated, however, to show that recovery is very seldom a simple process. Actually, if the equilibrium concentration of vacancies at the quench temperature is large, divacancies are very likely to be formed during quench, which influence the annealing kinetics.[2,3] Moreover, electron microscope observations by Silcox and Hirsch[4] and investigation of changes in mechanical properties during annealing by Mori, Meshii, and Kauffman[5] provided clear experimental evidence that the formation of vacancy clusters is also important in quenched gold.

Annealing kinetics seem to be simple and reflect essentially the behavior of pure vacancies only if the

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