of the inelastic scattering for momentum transfer equal to k_0 can be used. An analysis of such measurements in terms of a Lorentzian shape with energy half-width Γ has been reported[5] for $T > T_N$. As remarked in Sec. II, this approximation to the line shape does not yield a convergent integral for $\overline{\Omega}$ in Eq. (11). If the measurement at $T-T_N = 8$ K is used with a Lorentzian cutoff at energy transfer 5Γ, the result is $\overline{\Omega} = 7.2\times 10^{-3}$. If the cutoff is at 10Γ, the result is $\overline{\Omega} = 1.6\times 10^{-2}$. Even with the larger cutoff, the maximum possible difference here between χ_T and $\beta S^{(0)}$ is only 0.14%. The experimental width Γ decreases on further approach to T_N, and the possible deviation between χ_T and $\beta S^{(0)}$ also decreases.

A more precise application of such measurements to this topic and to the determination of the second moment $F^{(2)}$ will depend on improved measurements of the spectral shape function at large energy transfers.

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General Lattice Model of Phase Transitions

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A general lattice-statistical model which includes all soluble two-dimensional model of phase transitions is proposed. Besides the well-known Ising and "ice" models, other soluble cases are also considered. After discussing some general symmetry properties of this model, we consider in detail a particular class of the soluble cases, the "free-fermion" model. The explicit expressions for all thermodynamic functions with the inclusion of an external electric field are obtained. It is shown that both the specific heat and the polarizability of the free-fermion model exhibit in general a logarithmic singularity. An inverse-square-root singularity results, however, if the free-fermion model also satisfies the ice condition. The results are illustrated with a specific example.

I. INTRODUCTION

Considerations of the phenomena of phase transitions have been, to a large extent, centered around the study of lattice systems. Besides the intrinsic interest surrounding the lattice systems as models of real physical situations, one is further attracted to their consideration by the possibility of obtaining exact nontrivial solutions. But the soluble problems are very few in number. The Ising model[1,2] of magnetism, first proposed some

40 years ago, still stands at the very frontiers of present knowledge. The only other nontrivial models of phase transitions possessing rigorous solutions are the recently solved models of hydrogen-bonded ferroelectrics and antiferroelectrics.[3-9] It is perhaps not too surprising to find that, while the physical mechanisms responsible for the phase changes associated with the Ising and the ferroelectric models are quite distinct, the mathematical descriptions of these models are not too different. The central mathematical problem involved in all these models is to evaluate a certain generating function in the language of linear graphs. In an effort to search for further soluble problems, we have previously extended these considerations by proposing a general lattice-statistical problem.[9] While on the one hand this problem appears as a general model of ferroelectrics, including all the previously solved models, on the other, it also includes a number of yet unsolved statistical problems. In Ref. 9, we considered an approximate treatment of one aspect of the unsolved problems, namely, the next-neighbor Ising problem. However, there exist other soluble cases of this general problem which do not correspond to any of the known solutions that have been hitherto discussed. In the present paper we return to the study of these situations. The problem under consideration is first defined in Sec. II. Some general symmetry properties of the model are considered in Sec. III where the soluble models are categorized. The thermodynamic properties of one such category, the free-fermion model, are discussed in detail in Sec. IV. The results are illustrated by a specific example in Sec. V.

II. DEFINITION OF PROBLEM

Consider a periodic square lattice composed of N lattice sites (or vertices) and of $2N$ lattice edges. An edge can be either covered by a bond or empty. A definite covering of the lattice edges is called a bond complection G. Clearly there are a total of 2^{2N} distinct bond complections. The number of bonds incident to a vertex in a given bond complection is the degree of the vertex. A bond complection will in general consist of vertices of degrees ranging from 0 to q, where q is the coordination number of the lattice ($q=4$ for square lattice). We shall confine ourselves to considerations of bond complections G' consisting of vertices of *even* degrees only. There are then eight different types of bond configurations which may appear at a vertex. These are numbered from 1 to 8 as shown in Fig. 1. An empty lattice edge will be called a hole. Two bond configurations (or complections) are conjugate to each other if they are related through the interchange of all holes and bonds. Thus, for instance, the vertex types (1) and (2) of Fig. 1 are mutually conjugate. Next, a weight factor $\omega(\xi)$ is associated with each vertex configuration of type $\xi(=1, 2, \ldots, 8)$. The weight W_G of a bond complection G is then taken to be the product of all N vertex weights. The mathematical problem we face is to evaluate the partition sum or the generating function

$$Z=\sum_{G'} W_{G'}=\sum_{G'}\prod_{i=1}^{N}\omega(\xi_i)\,. \tag{1}$$

Here, the summation is extended over all bond complections G' consisting of vertices of even degrees; the symbol ξ_i refers to the type of configuration at the ith vertex for a given bond complection G'.

For problems of physical interest, energies are assigned to the different vertex configurations and the weights $\omega(\xi)$ are simply the Boltzmann factors

$$\omega(\xi)=\exp(-\beta e_\xi)\,, \tag{2}$$

where e_ξ is the energy assigned to the ξth type of vertex configuration, $T=(k\beta)^{-1}$ is the temperature, and k is the Boltzmann constant. In such cases, Z is the partition function of the system and the thermodynamic functions can be deduced from the free energy per vertex

$$f=-\beta^{-1}\lim_{N\to\infty}\frac{1}{N}\ln Z. \tag{3}$$

The previous models of phase transitions are recovered for special choices of the vertex energies. For easy reference, a collection of these specializations is included in Table I.

One physical quantity of interest in the consideration of the ferroelectric and antiferroelectric models is the polarization. In these models, each lattice edge is considered to carry a (unit) dipole moment. We now make the correspondence that an edge covered by a bond means a dipole pointing toward the left (for horizontal bonds) or in the upward direction (for vertical bonds). Then, in the presence of an external field $\vec{\mathcal{E}}=(h, v)$, the dipole energies $-\vec{\mathrm{d}}\cdot\vec{\mathcal{E}}$ are included by redefining the vertex energies

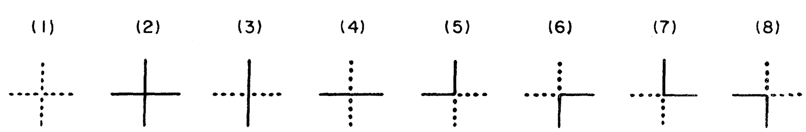

FIG. 1. The eight different kinds of vertex configurations.

TABLE I. Reduction of the general problem to soluble models.

	e_1	e_2	e_3	e_4	e_5	e_6	e_7	e_8	Singularity in specific heat
KDP[a]	0	0	ϵ	ϵ	ϵ	ϵ	∞	∞	$(T-T_c)^{-1/2}$
F[b]	ϵ	ϵ	ϵ	ϵ	0	0	∞	∞	none[c]
Modified KDP[d]	∞	ϵ_2	ϵ_3	ϵ_4	ϵ_5	ϵ_6	∞	∞	$(T-T_c)^{-1/2}$
Modified F[e]	ϵ	ϵ	ϵ	ϵ	0	0	2ϵ	2ϵ	$\ln\lvert T-T_c\rvert$
Rectangular Ising[f]	$-(J_1+J_2)$	J_1+J_2	J_1-J_2	$-J_1+J_2$	0	0	0	0	$\ln\lvert T-T_c\rvert$
Triangular Ising[g]	$-(J_1+J_2+J_3)$	$J_1+J_2-J_3$	$J_1-J_2+J_3$	$-J_1+J_2+J_3$	J_3	J_3	$-J_3$	$-J_3$	$\ln\lvert T-T_c\rvert$
Free-fermion Models[h]			$e_1+e_2=e_7+e_8$,	$e_3+e_4=e_5+e_6$					$\ln\lvert T-T_c\rvert$[h]
Conjugate Models[i]	δ	δ	$-\delta$	$-\delta$	$-\epsilon$	$-\epsilon$	$-\epsilon$	$-\epsilon$	$\ln\lvert T-T_c\rvert$

[a]Reference 3.
[b]Reference 3.
[c]A limiting situation of λ transitions, see Ref. 8.
[d]Reference 6, restricted to $\epsilon_3+\epsilon_4=\epsilon_5+\epsilon_6$.
[e]Reference 7. This is a special case of the conjugate model.
[f]Reference 9.
[g]The vertex energies are obtained by putting $J'=0$ in Eq. (A1) of Ref. 7. The resulting critical condition (34) for the triangular lattice now has a compact form and applies to both ferromagnetic and antiferromagnetic interactions.
[h]This becomes the modified KDP model if $e_1=\infty$.
[i]See description in text. Here we have taken $u_1u_2=u_3u_4=1$.

$$\begin{aligned}
&\bar{e}_1=e_1-(h+v), \qquad &&\bar{e}_5=e_5\,,\\
&\bar{e}_2=e_2+(h+v)\,, &&\bar{e}_6=e_6\,,\\
&\bar{e}_3=e_3-(h-v), &&\bar{e}_7=e_7\,,\\
&\bar{e}_4=e_4+(h-v), &&\bar{e}_8=e_8\,.
\end{aligned} \tag{4}$$

Consequently, the polarization $\vec{\mathrm{P}}$ is given by the expressions

$$P_x=-kT\frac{\partial\bar{f}}{\partial h}\,, \qquad P_y=-kT\frac{\partial\bar{f}}{\partial v}\,, \tag{5}$$

where $\bar{f}$ is the free energy (3) evaluated with the energies $\bar{e}_i$ in the place of e_i.

III. GENERAL CONSIDERATIONS

The partition function (1) has not been evaluated in a closed form for its most general expression with arbitrary vertex energies. The general partition function possesses, however, a number of symmetry properties that can be obtained through the following considerations.[10]

First, it is easy to see that pairs of vertices (5) and (6) or (7) and (8) occur together. Therefore, without loss of generality, we may take $\omega(5)=\omega(6)=a$, $\omega(7)=\omega(8)=b$. Consequently, Z is invariant under the change of sign of a or b. The left-right symmetry then leads to the symmetry relation

$$Z(a,b)=Z(b,a)\,. \tag{6}$$

We shall now temporarily disregard the weights a and b and write $Z=Z_{1234}$, where each numerical $\xi(=1,2,3,4)$ stands for the vertex weight $\omega(\xi)$. Since it is immaterial whether to call the bonds holes or bonds, Z is invariant under the interchange of bonds and holes in a given (vertical or horizontal) direction or in both directions simultaneously. We then have the symmetry relation[10]

$$Z=Z_{1234}=Z_{2143}=Z_{4321}=Z_{3412}\,. \tag{7a}$$

Furthermore, a 90° rotation of the lattice interchanges only the indices 3 and 4 (and also the symmetric weights a and b); hence we have

$$Z=Z_{1243}=Z_{2134}=Z_{4312}=Z_{3421}\,. \tag{7b}$$

Relations (6) and (7) tell us that the partition function is invariant with respect to interchanges between conjugate pairs provided that no changes between the vertices 1, 2, 3, 4 and 5, 6, 7, 8 occur.

If the conjugate pairs have the same weights, further symmetry relations exist which permit permutations between the vertices 1, 2, 3, 4 and 5, 6, 7, 8. Let us denote the weights by

$$\omega(1)=\omega(2)=u_1, \qquad \omega(3)=\omega(4)=u_2\,,$$

$$\omega(5)=\omega(6)=u_3, \qquad \omega(7)=\omega(8)=u_4, \tag{8}$$

and write $Z=Z(u_1, u_2; u_3, u_4)$.

Equations (6) and (7) now read

$$Z=Z(u_2, u_1; u_3, u_4)=Z(u_1, u_2; u_4, u_3)\,. \tag{9}$$

We now interchange the bonds and holes along the zigzag paths shown in Fig. 2. Decomposing the lattice into two interconnecting sublattices A and B, it is then easy to see that if on sublattice A we have the following vertex interchanges:

$$\begin{aligned}&(1)\leftrightarrow(6), \quad (2)\leftrightarrow(5),\\ &(3)\leftrightarrow(7), \quad (4)\leftrightarrow(8),\end{aligned} \tag{10a}$$

then on sublattice B we have the interchanges

$$\begin{aligned}&(1)\leftrightarrow(5), \quad (2)\leftrightarrow(6),\\ &(3)\leftrightarrow(8), \quad (4)\leftrightarrow(7)\,.\end{aligned} \tag{10b}$$

Using (8), we see that (10a) and (10b) are identical and we are led to the further relation

$$Z\equiv Z(u_1, u_2; u_3, u_4)=Z(u_3, u_4; u_1, u_2)\,. \tag{11}$$

In both (9) and (11), it should be remembered that Z is also invariant under the replacement of any u_i by $-u_i$.

Another useful relation can be obtained by applying to the partition function a rearrangement procedure, known as the method of weak-graph expansion.[11] The discussion of the method and its application to the present problem have been previously given by one of us.[7] We write here only the resulting relation[12]

$$\begin{aligned}Z(u_1, u_2; u_3, u_4)&\\ &=Z\big(\tfrac{1}{2}(u_1+u_2+u_3+u_4), \tfrac{1}{2}(u_1+u_2-u_3-u_4);\\ &\qquad \tfrac{1}{2}(u_1-u_2+u_3-u_4), \tfrac{1}{2}(u_1-u_2-u_3+u_4)\big)\,.\end{aligned} \tag{12}$$

Further iterations of (12) yield no new relations except those specified by (9) and (11) and the replacement of u_i by $-u_i$.

The reduction of this general problem into specific models has been given in Table I. For some of these special cases, a closed form of the free energy can be obtained. We classify these soluble cases into the following categories.

a. The ice models.[3–6] Included in this category are the models with the ice-condition constraint $\omega(7)=\omega(8)=0$ or equivalently $e_7=e_8=\infty$. These include the potassium dihydrogen phosphate KDP model of ferroelectrics and the F model of antiferroelectrics considered by Lieb.[3] The exact solution of the ice models can be obtained by a method which explicitly uses the fact that the vertices (7) and (8) are excluded.[13] The readers are referred to Refs. 3 and 4 for detailed discussions of these solutions. The specific heat exhibits in general a $|T-T_c|^{-1/2}$ singularity in the ice models.[14]

b. The conjugate models. Included in this category are the models specified by (8) but with the further constraint

$$u_1u_2=u_3u_4\,. \tag{13}$$

It has been shown[15] that this model is equivalent to an Ising model of a rectangular lattice with the interactions $J=\frac{1}{2}(e_1-e_3)$ and $J'=\frac{1}{2}(e_2-e_3)$. Hence, the specific heat exhibits a $\ln|T-T_c|$ singularity. The free energy has the following closed expression[12]:

$$\begin{aligned}\beta f=-\frac{1}{8\pi^2}\int_0^{2\pi}d\theta\int_0^{2\pi}d\phi\, \ln[&(u_1+u_2)^2+(u_3-u_4)^2\\ &+2(u_1-u_3)(u_2-u_4)\cos(\theta-\phi)\\ &+2(u_1-u_4)(u_2-u_3)\cos(\theta+\phi)]\,.\end{aligned} \tag{14}$$

This expression can also be obtained by using (12) to convert the model into the free-fermion model considered in the following. An interesting application is the modified F model of antiferroelectrics considered by one of us.[7]

c. The free-fermion models. Included in this category are the models satisfying the relation[16]

$$\omega(1)\omega(2)+\omega(3)\omega(4)=\omega(5)\omega(6)+\omega(7)\omega(8), \tag{15}$$

which we refer to as the free-fermion condition. This category includes the various planar Ising

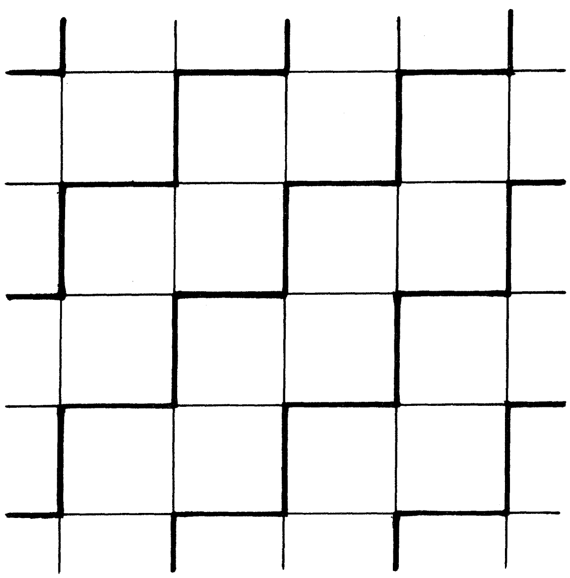

FIG. 2. The zig-zag paths along which the bonds and holes are interchanged to obtain the symmetry relation (11).

models and the modified KDP model of ferroelectrics.[5,6] A closed expression for the free energy, which is valid when (15) holds, has been obtained in Ref. 9 using a method similar to the S-matrix formulation for the many-body problem. It was seen there that the imposition of the condition (15) is equivalent to the consideration of a system of noninteracting fermions. For completeness, we now give below the closed expression for the free energy f and include in the Appendix an alternative derivation using the method of dimers:

$$\beta f = -\frac{1}{8\pi^2}\int_0^{2\pi} d\theta \int_0^{2\pi} d\phi \ln[2a + 2b\cos\theta + 2c\cos\phi + 2d\cos(\theta-\phi) + 2e\cos(\theta+\phi)], \tag{16}$$

where

$$\begin{aligned} 2a &= [\omega(1)]^2 + [\omega(2)]^2 + [\omega(3)]^2 + [\omega(4)]^2, \\ b &= \omega(1)\omega(3) - \omega(2)\omega(4), \\ c &= \omega(1)\omega(4) - \omega(2)\omega(3), \\ d &= \omega(3)\omega(4) - \omega(7)\omega(8), \\ e &= \omega(3)\omega(4) - \omega(5)\omega(6). \end{aligned} \tag{17}$$

It is to be noted that the free-fermion condition (15) remains as an identity when we replace e_i by $\bar{e}_i$ as given by (4). Hence, it is possible to discuss the properties of the free-fermion model under an external electric field. As seen from Table I, special cases of the free-fermion model include the various planar Ising models as well as the modified KDP model whose thermodynamic properties are well known. Besides these special cases, however, discussions of the thermodynamic properties of the free-fermion models in general have not been given.[17] Such a study is especially useful in view of the different critical behaviors exhibited by the Ising and the modified KDP models. For example, it would be illuminating to see how the change of critical behavior comes about as the vertex energies are varied. These discussions will be the subject of Sec. IV.

IV. FREE-FERMION MODEL

In this section we discuss the thermodynamic properties of the free-fermion model defined by (15). For the free-fermion condition (15) to hold at all temperatures, we must have the identities

$$\{e_1+e_2,\ e_3+e_4\} = \{e_5+e_6,\ e_7+e_8\}. \tag{18}$$

Because of the symmetry relation (6), we may take, without loss of generality,

$$e_1+e_2 = e_7+e_8, \quad e_3+e_4 = e_5+e_6. \tag{19}$$

Then, the free energy is given only in terms of ω_1, ω_2, ω_3, and ω_4. Writing $\omega_i \equiv \omega(i)$, we have explicitly

$$\begin{aligned} a &= \tfrac{1}{2}(\omega_1^2+\omega_2^2+\omega_3^2+\omega_4^2), \quad b = \omega_1\omega_3 - \omega_2\omega_4, \\ c &= \omega_1\omega_4 - \omega_2\omega_3, \quad d = \omega_3\omega_4 - \omega_1\omega_2, \quad e = 0. \end{aligned} \tag{20}$$

One of the two integrations in (16) can be performed. Here, we use the integration formula

$$\int_0^{2\pi} d\theta \ln(2A + 2B\cos\theta + 2C\sin\theta) = 2\pi \ln[A + (A^2 - B^2 - C^2)^{1/2}], \tag{21}$$

where for our purposes

$$A = a + c\cos\phi, \quad B = b + d\cos\phi, \quad C = -d\sin\phi. \tag{22}$$

Then, expressions (16) and (21) lead to the following expression:

$$\beta f = -\frac{1}{4\pi}\int_0^{2\pi} d\phi \ln\{A + [Q(\phi)]^{1/2}\}. \tag{23}$$

Here

$$\begin{aligned} Q(\phi) &= A^2 - B^2 - C^2 = y^2 + z^2 - x^2 - 2yz\cos\phi + x^2\cos^2\phi \\ &= x^2(\cos\phi - yz/x^2)^2 + (1/x^2)(x^2 - y^2)(z^2 - x^2), \end{aligned} \tag{24}$$

and

$$x = \omega_2\omega_3 - \omega_1\omega_4, \quad y = \tfrac{1}{2}(\omega_1^2 - \omega_2^2 - \omega_3^2 + \omega_4^2), \quad z = \omega_2\omega_3 + \omega_1\omega_4. \tag{25}$$

Clearly, in the discussion of the analytic properties of f, it is important to consider whether $Q(\phi)$ is a complete square. We have the following cases to consider.

Case (1): *$Q(\phi)$ is a complete square*. These are two possibilities.

(a) $x^2 = z^2$: This is equivalent to the condition $\omega_1\omega_2\omega_3\omega_4 = 0$. Without loss of generality, we may take $\omega_1 = 0$. Then, the free-fermion condition (19) also implies $\omega_7\omega_8 = 0$ and the problem reduces to a special case of the ice models, i.e., the modified KDP model considered in Ref. 6. More explicitly, (23) becomes

$$\beta f = -\frac{1}{4\pi}\int_0^{2\pi} d\phi \ln\tfrac{1}{2}(\omega_2^2 + \omega_3^2 + \omega_4^2 - 2\omega_2\omega_3\cos\phi + |\omega_2^2 + \omega_3^2 - \omega_4^2 - 2\omega_2\omega_3\cos\phi|), \tag{26}$$

which is precisely Eq. (7) of Ref. 6. The important thermodynamic properties of this model are summarized as follows. A second-order phase transition occurs at a temperature T_c determined by

$$\omega_2 + \omega_3 + \omega_4 = 2\max\{\omega_2, \omega_3, \omega_4\}. \tag{27}$$

The specific heat c vanishes for $T < T_c$ and behaves as $(T - T_c)^{-1/2}$ near and above T_c. We note that we have obtained here the $(T - T_c)^{-1/2}$ singularity of

the ice models.

(b) $x^2=y^2$: This is equivalent to the condition

$$(\omega_1+\omega_2-\omega_3-\omega_4)(\omega_1-\omega_2-\omega_3+\omega_4)\times(\omega_1-\omega_2+\omega_3-\omega_4)=0. \tag{28}$$

We then have two alternatives if (28) is to hold at all temperatures. (i) $\{e_1, e_2\}=\{e_3, e_4\}$. It is easy to see that (23) reduces to

$$\beta f=-\tfrac{1}{2}\ln(2(\omega_1^2+\omega_2^2)). \tag{29}$$

(ii) $\{e_1, e_3\}=\{e_2, e_4\}$. Again we have the simple expression

$$\beta f=-\tfrac{1}{2}\ln(2(\omega_1^2+\omega_3^2+\omega_1\omega_3)). \tag{30}$$

In either case, the model exhibits no phase transition, a fact which can also be seen from the fact that the ground state is macroscopically degenerate.

Case (2): $Q(\phi)$ *is not a complete square*. One observes from (23) that f is analytic in T except at the points given by $Q(\phi)=0$, where some derivatives of f diverge. Now we find $Q(\phi)\geq 0$ by (24). Therefore, the critical condition is obtained by setting the absolute minimum of $Q(\phi)$ equal to zero. Since

$$Q'(\phi)=2x^2\sin\phi\,(yz/x^2-\cos\phi), \tag{31}$$

there are three possible extreme values for $Q(\phi)$,

$$Q(0)=(y-z)^2, \quad Q(\pi)=(y+z)^2, \quad Q(\phi_0)=(1/x^2)(x^2-y^2)(z^2-x^2)>0, \tag{32}$$

where $\cos\phi_0=yz/x^2$ for $|yz|\leq x^2$, and $Q(\phi_0)$ is positive because $z^2\geq x^2$, and hence $x^2\geq y^2$. Therefore, the only possibilities are to set $Q(0)$ or $Q(\pi)$ equal to zero. This is equivalent to $z^2=y^2$ and hence to the critical condition

$$(-\omega_1+\omega_2+\omega_3+\omega_4)(\omega_1-\omega_2+\omega_3+\omega_4)(\omega_1+\omega_2-\omega_3+\omega_4)(\omega_1+\omega_2+\omega_3-\omega_4)=0. \tag{33}$$

Equivalently, (33) can be rewritten as

$$\omega_1+\omega_2+\omega_3+\omega_4=2\max\{\omega_1, \omega_2, \omega_3, \omega_4\}. \tag{34}$$

It is now straightforward, although tedious, to compute the energy E through direct differentiation of (23): We find

$$E=\frac{\partial}{\partial\beta}(\beta f)=-\frac{1}{4\pi}\int_0^{2\pi}d\phi\left[\frac{BB'+CC'}{B^2+C^2}+\left(A'-A\frac{BB'+CC'}{B^2+C^2}\right)[Q(\phi)]^{-1/2}\right], \tag{35}$$

where the primes denote derivatives with respect to β. We now introduce (22) and after some reduction we arrive at the expression

$$E=C_0+C_1I_1+C_2I_2+C_3I_3, \tag{36}$$

where

$$C_0=-\frac{1}{4\pi}\int_0^{2\pi}d\phi\left(\frac{BB'+CC'}{B^2+C^2}\right)$$
$$=-\frac{b'}{2b} \quad \text{if } b^2>d^2 \quad \text{or } \operatorname{sgn}(e_1-e_4)=\operatorname{sgn}(e_3-e_2),$$
$$=-\frac{d'}{2d} \quad \text{if } b^2<d^2 \quad \text{or } \operatorname{sgn}(e_1-e_4)=\operatorname{sgn}(e_2-e_3),$$

$$C_1=-a'+\frac{a}{2}\left(\frac{b'}{b}+\frac{d'}{d}\right)+\frac{c}{4bd}\left(\frac{b'}{b}-\frac{d'}{d}\right)(b^2-d^2), \tag{37}$$
$$C_2=-c'+\frac{c}{2}\left(\frac{b'}{b}+\frac{d'}{d}\right),$$
$$C_3=\frac{1}{2}\left(\frac{b'}{b}-\frac{d'}{d}\right)(b^2-d^2)\left(\frac{a}{b^2+d^2}-\frac{c}{2bd}\right),$$

and I_1, I_2, and I_3 are definite integrals given by

$$I_1=(4\pi)^{-1}\int_0^{2\pi}[Q(\phi)]^{-1/2}d\phi,$$

$$I_2=(4\pi)^{-1}\int_0^{2\pi}\cos\phi[Q(\phi)]^{-1/2}d\phi, \tag{38}$$

$$I_3=(4\pi)^{-1}\int_0^{2\pi}(1+\omega\cos\phi)^{-1}[Q(\phi)]^{-1/2}d\phi,$$

$$\omega=2bd/(b^2+d^2).$$

In all these formulas, $Q(\phi)$ is given by (24) and a, b, c, d given by (20). The integrals I_1, I_2, and I_3 can be expressed in terms of the complete elliptical integrals of the first and the third kinds. In evaluating these integrals, it is sufficient to consider positive x, y, and z only. For with negative x, y, or z, we may use the simple relations

$$I_1(x, y, z)=I_1(\pm x, \pm y, \pm z),$$
$$I_2(x, -y, z)=-I_2(x, y, z),$$
$$I_3(x, -y, z, \omega)=I_3(x, y, z, -\omega), \text{ etc.}$$

For positive x, y, z, the following definitions of a new variable α are useful:

$$z>x>y>0, \quad \cos\alpha=(z\cos\phi-y)/(z-y\cos\phi),$$
$$z>y>x>0, \quad \sin\alpha=(z\cos\phi-y)/(z-y\cos\phi), \tag{39}$$
$$y>z>x>0, \quad \sin\alpha=(y\cos\phi-z)/(y-z\cos\phi).$$

It is then elementary to find

(i) $z>x>y>0$:

$$I_1=\frac{1}{\pi(z^2-y^2)^{1/2}}K(k),$$

$$I_2=\frac{z}{\pi y(z^2-y^2)^{1/2}}[K(k)-\Pi(n, k,)],$$

$$I_3=\frac{1}{\pi(y+\omega z)(z^2-y^2)^{1/2}}\times\left(yK(k)+\frac{\omega(z+\omega y)}{1-\omega^2}\,\Pi(m,k)\right),$$

$$\text{where}\quad k^2=(x^2-y^2)/(z^2-y^2),\quad n=y^2/(z^2-y^2),\quad m=(\omega y+z)^2/(1-\omega^2)(z^2-y^2). \tag{40}$$

(ii) $z>y>x>0$:

$$I_1=\frac{1}{\pi(z^2-x^2)}K(g),$$

$$I_2=\frac{1}{\pi y(z^2-x^2)^{1/2}}\,[z^2K(g)+(y^2-z^2)\,\Pi(r,k)]\,,$$

$$I_3=\frac{1}{\pi(y+\omega z)(z^2-x^2)^{1/2}}\left(gK(g)+\frac{\omega(z^2-y^2)}{z+\omega y}\,\Pi(s,k)\right).$$

$$\text{where}\quad q^2=(y^2-x^2)/(z^2-x^2)\,,\quad r=-y^2/z^2,\quad s=-(\omega z+y)^2/(z+\omega y)^2. \tag{41}$$

(iii) $y>z>x>0$: Same as case (ii) only with y and z interchanged. In these expressions, K and Π are, respectively, the complete elliptical integrals of the first and the third kinds defined by

$$K(k)=\int_0^{\pi/2}(1-k^2\sin^2\alpha)^{-1/2}\,d\alpha,$$
$$\Pi(n,k)=\int_0^{\pi/2}(1+n\sin^2\alpha)^{-1}(1-k^2\sin^2\alpha)^{-1/2}\,d\alpha. \tag{42}$$

The phase transition occurs at $|y|=|z|$ or $q=1$, $r=-1$. While K and Π diverge as $\ln|T-T_c|$ when $q\to 1$, it can be seen from (36) and (37) that

$$E\to C_0+|T-T_c|\ln|T-T_c|\,.$$

Hence, E is continuous at T_c and the specific heat diverges logarithmically near T_c.

A special feature is that the energy E has different expressions for $|x|>|y|$ and $|x|<|y|$, although $|x|=|y|$ is not a singular point. Since for the quartic Ising model only one expression is needed for E, it is of interest to investigate the condition under which $|x|=|y|$ may occur. To obtain such a criterion, let us arrange the energy values e_1, e_2, e_3, and e_4 according to their magnitudes such that

$$e_\alpha\ge e_\beta\ge e_\gamma\ge e_\delta\,.$$

Then, $x^2=y^2$ corresponds to

$$\omega_\alpha+\omega_\delta=\omega_\beta+\omega_\gamma\,, \tag{43}$$

where $\omega_\alpha=\exp(-e_\alpha/kT)$, etc. Equation (43) can be rewritten as

$$\exp\left\{\frac{1}{2kT}\,[(e_\beta+e_\gamma)-(e_\alpha+e_\delta)]\right\}=\cosh\left(\frac{e_\alpha-e_\delta}{2kT}\right)\Big/\cosh\left(\frac{e_\beta-e_\gamma}{2kT}\right)>1\,.$$

Hence, (43) has a solution, and thus it is possible to have $x^2=y^2$ only for

$$e_\beta+e_\gamma<e_\alpha+e_\delta\ .$$

It is not difficult to show that, in fact, $|x|>|y|$ at all temperatures if $e_\beta+e_\gamma\ge e_\alpha+e_\delta$. An example is the quartic Ising model for which the equality holds (cf. Table I), and the energy is given by the same single expression both above and below the transition temperature T_c. For a triangular Ising lattice with the corresponding vertex energies given in Table I, it is possible to satisfy (43) whenever the interactions are antiferromagnetic. The temperature thus determined is the disorder temperature[18] at which the spin correlations undergo a remarkable change.[19]

Finally, we consider the inclusion of an external field $\vec{\mathcal{E}}$. Earlier, we have shown that the inclusion of a field amounts simply to redefining the vertex energies. Since the new vertex energies (4) also happen to satisfy the free-fermion condition, we have already solved the problem. In fact, all analyses of this section again go through provided that we use the new vertex energies in all formulas. In particular, the transition temperature T_c now depends on the external field and is determined by

$$\omega_1HV+\omega_2(HV)^{-1}+\omega_3HV^{-1}+\omega_4H^{-1}V=2\max\{\omega_1HV,\omega_2(HV)^{-1},\omega_3HV^{-1},\omega_4H^{-1}V\}\,, \tag{44}$$

where $H\equiv e^{\beta h}$, $V\equiv e^{\beta v}$.

The polarizations $\vec{\mathrm{P}}=(P_x, P_y)$ for the free-fermion model can be obtained from (5) and (23). The expression for P_x is given below in (45) and (46), while the expression for P_y is the same except for the interchange of ω_3 and ω_4:

$$P_x=\frac{2}{\pi}\frac{z^2-y^2}{y(z^2-x^2)^{1/2}}[\Pi(n,k)-K(k)],\qquad T>T_c$$
$$P_x=\frac{2}{\pi}\frac{y^2-z^2}{y(y^2-x^2)^{1/2}}\,\Pi(n,k),\qquad T<T_c. \tag{45}$$

Near T_c, we have asymptotically

$$P_x=\frac{z}{x}\left(1-\frac{2}{\pi}\cos^{-1}\frac{x}{z}+\frac{x(z^2-y^2)}{2\pi z^2(z^2-x^2)^{1/2}}\right)+\frac{y^2-z^2}{\pi z(z^2-x^2)^{1/2}}\ln\frac{4}{k'}+O(k'^4),\qquad T>T_c$$

$$P_x=\frac{z}{x}\left(1-\frac{2}{\pi}\cos^{-1}\frac{x}{y}+\frac{y^2-z^2}{2\pi y^2(y^2-x^2)^{1/2}}\right) \tag{46}$$

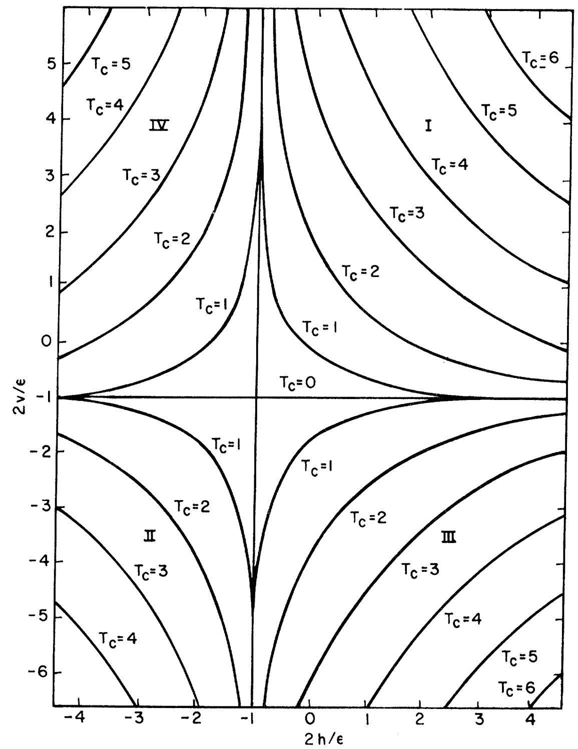

FIG. 3. Constant T_c contours in the hv plane. The transition temperature T_c is measured in units ϵ/k.

$$+\frac{z^2-y^2}{\pi y(y^2-x^2)^{1/2}}\ln\frac{4}{k'}+O(k'^4)\,, \qquad T<T_c$$

where $k'=(1-k^2)^{1/2}$. The polarization is nonvanishing and continuous at all temperatures. It behaves near T_c as $C_0+C_1(T-T_c)\ln(T-T_c)$. Therefore, the polarizability $\partial\vec{P}/\partial\vec{\mathcal{E}}$ exhibits a logarithmic singularity. Thus, we see that in the ferroelectric version of the free-fermion model, the phase transition is not marked by the appearance or vanishing of the order-parameter polarization. It is instead associated with the logarithmic divergence in the specific heat and the polarizability. We also note that none of these thermodynamic functions has a simple power-law dependence on $|T-T_c|$ near T_c.

V. NUMERICAL EXAMPLE

To illustrate the above results, we shall now consider a specific example with vertex energies

$$\begin{aligned} &e_1=0, \qquad e_2=2\epsilon>0\,, \\ &e_3=e_4=e_5=e_6=e_7=e_8=\epsilon\,. \end{aligned} \tag{47}$$

As seen from Table I, this specifies precisely the regular Ising model with an interaction parameter $J_1=J_2=\frac{1}{2}\epsilon$. A plot of the energy and the specific heat of this model can be found in Ref. 7 and will not be reproduced here.[20] The transition temperature is given by $kT_c/\epsilon=\ln(\sqrt{2}+1)\equiv 1.13459\ldots$. We shall now consider the ferroelectric version of the model by imposing an external electric field. The transition temperature T_c is now given by

$$\begin{aligned} &HV+(HV)^{-1}u^2+uHV^{-1}+uH^{-1}V \\ &\qquad =2\max\{HV,(HV)^{-1}u^2,uHV^{-1},uH^{-1}V\}, \end{aligned} \tag{48}$$

where $u\equiv e^{-\beta\epsilon}$.

The entire hv plane is then divided into four regions (see Fig. 3) depending on which vertex 1, 2, 3, or 4 has the lowest energy (or the largest weight). We have plotted in Fig. 3 the constant T_c contours in the hv plane. The four regions denoted by I, II, III, and IV are, respectively, the regions where the vertex 1, 2, 3, or 4 has lowest energy. Relating the vertex energies – including the external field (h,v) – to the interactions J_1 and J_2 of a rectangular Ising lattice (see Table I), it can be seen that our model is equivalent to rectangular Ising lattice with $J_1=\frac{1}{2}\epsilon+v$, $J_2=\frac{1}{2}\epsilon+h$. Hence (48) is equivalent to the well-known relation

$$\left|\sinh\frac{\epsilon+2v}{2kT_c}\,\sinh\frac{\epsilon+2h}{2kT_c}\right|=1. \tag{49}$$

For $v=0$, T_c as a function of h is plotted in Fig. 4. Finally, we plot in Fig. 5 the polarization P_x as a function of T.

VI. CONCLUSIONS

We have defined a general lattice-statistical problem and studied in detail the soluble situations. It is found that the specific heat behaves as $|T-T_c|^{-1/2}$ near T_c for the ice models and possesses the $\ln|T-T_c|$ singularity for all other soluble cases. From these results and the study of related models,[7,8] it appears that the logarithmic singularity is perhaps commonplace except in the ice models, which happen to exhibit a $|T-T_c|^{-1/2}$ singularity in the specific heat.

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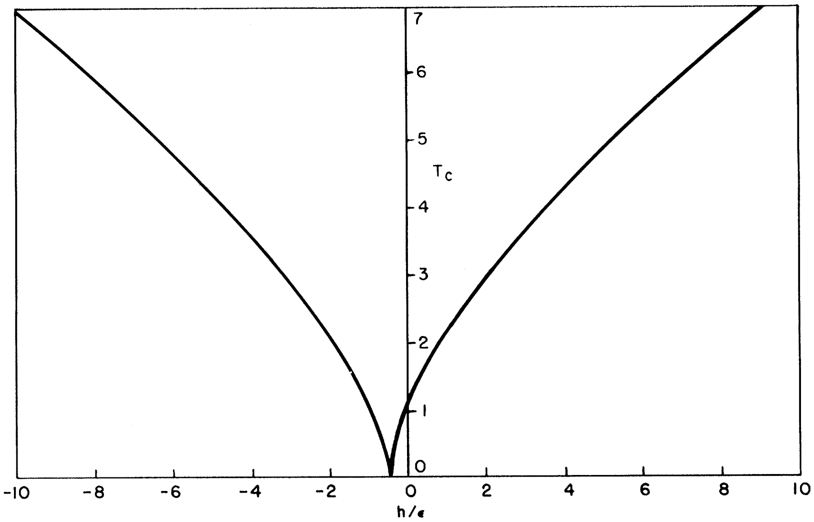

FIG. 4. The transition temperature T_c as a function of h with $v=0$. T_c is in units ϵ/k.

APPENDIX

In this Appendix, we derive the partition function (16) for the free-fermion models using the method of dimers.[21] First, we construct a dimer lattice by expanding each vertex of the square lattice into a city of internally connected points. Provided that the structure of the city and the edge weights are chosen properly, the partition function is identical to the generating function for closely packed dimer configurations on this expanded lattice. It turns out that this trick can be accomplished by choosing the *planar* dimer city of Fig. 6 with the weights shown in the graph.[22] In Fig. 7, we list all possible dimer configurations for a dimer city. It is then easy to see that the correct vertex weights are indeed generated. For example, the first row of Fig. 7 yields

$$\frac{\omega_7}{\omega_2}\times\omega_2\times\frac{\omega_8}{\omega_2}+\frac{\omega_6-\omega_4}{\omega_2}\times\omega_2\times\frac{\omega_5-\omega_3}{\omega_2}+\frac{\omega_6-\omega_4}{\omega_2}\times\omega_3\times 1$$
$$+\frac{\omega_5-\omega_3}{\omega_2}\times\omega_4\times 1=\frac{1}{\omega_2}(\omega_5\omega_6+\omega_7\omega_8-\omega_3\omega_4)=\omega_1 .$$

The last equality follows from the free-fermion condition (15). The other vertex weights can be generated accordingly.

The next step is to direct the edges of the dimer lattice so that every closed polygon drawn on the expanded dimer lattice containing an even number of edges and enclosing an even number of points has odd numbers of arrows pointing in each direc-

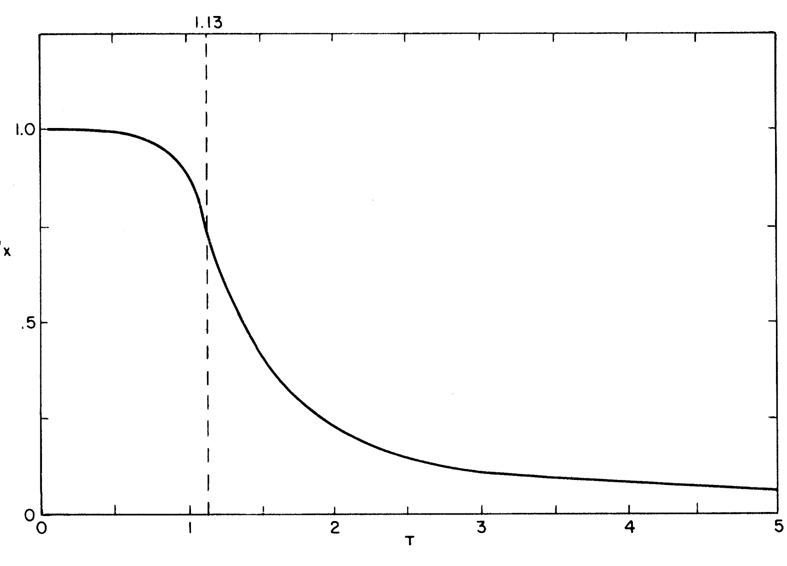

FIG. 5. Horizontal polarization as a function of temperature.

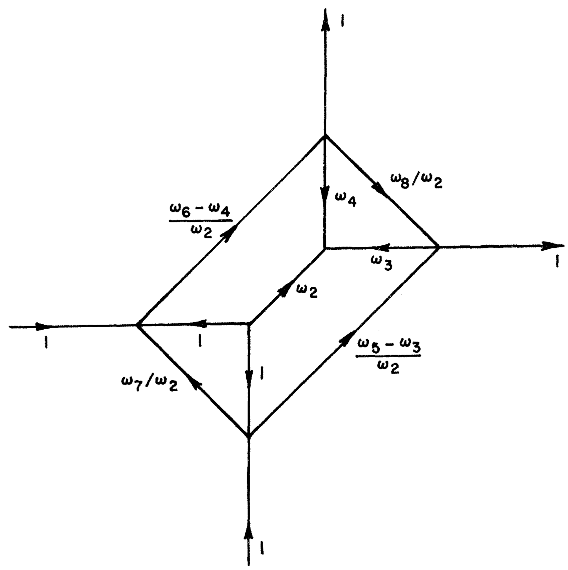

FIG. 6. The dimer city and the edge weights.

tion. This orientation can be accomplished as shown in Fig. 6. The dimer generating function, which is equal to the partition function, can now be written as a Pfaffian and evaluated accordingly.[21] For a periodic lattice, the result obtained is

$$\beta f=-\lim_{N\to\infty}\frac{1}{N}\ln Z=\frac{-1}{8\pi^2}\int_0^{2\pi}d\theta\int_0^{2\pi}d\phi\,\ln D, \qquad \text{(A1)}$$

where D is the determinant given by

$$D=\begin{vmatrix} 0 & \frac{\omega_8}{\omega_2} & 0 & \frac{\omega_4-\omega_6}{\omega_2} & e^{i\theta} & \omega_4 \\ -\frac{\omega_8}{\omega_2} & 0 & 0 & e^{i\phi} & \frac{\omega_3-\omega_5}{\omega_2} & \omega_3 \\ 0 & 0 & 0 & 1 & 1 & \omega_2 \\ \frac{\omega_6-\omega_4}{\omega_2} & -e^{-i\phi} & -1 & 0 & -\frac{\omega_7}{\omega_2} & 0 \\ -e^{-i\theta} & \frac{\omega_5-\omega_3}{\omega_2} & -1 & \frac{\omega_7}{\omega_2} & 0 & 0 \\ -\omega_4 & -\omega_3 & -\omega_2 & 0 & 0 & 0 \end{vmatrix}. \qquad \text{(A2)}$$

Substitution of (A2) into (A1) now yields (16).[23]

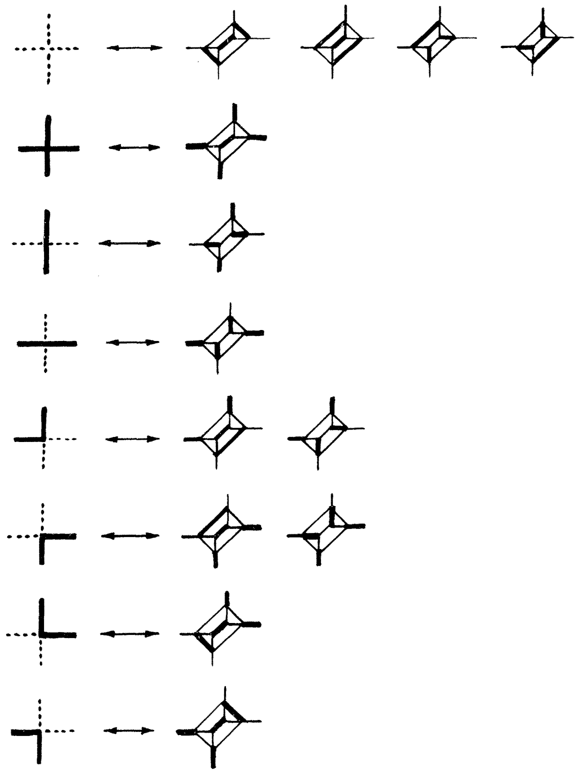

FIG. 7. The correspondences between the bond configurations and the dimer configurations.

Note added in proof. We now realize that case (2) of the free-fermion model discussed in Sec. IV is completely equivalent to the Ising model on an anistotropic triangular lattice. This makes our derivation of the logarithmic singularity in the specific heat obsolete. The formulas (31)–(40) are still useful, however, since none has been given in detail in the literature for the triangular Ising lattice.

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[12]Discussion in Ref. 7 was specialized to the case $u_3 = 1$, $u_4 = u_1 u_2$. The derivation of (12) and (14) presents no problem if one follows the procedures of Ref. 7 and uses the notations u_3 and u_4 in places of 1 and $u_1 u_2$.

[13]E. H. Lieb, Phys. Rev. Letters 18, 692 (1967).

[14]Except the F model for which the specific heat is continuous at T_c. But this has been identified in Ref. 8 as a limiting situation of the more familiar λ transitions.

[15]See the Appendix of Ref. 7.

[16]In the discussion of the general planar Ising models, Hurst and Green [H. S. Green and C. A. Hurst, in *Order-Disorder Phenomena*, edited by I. Preigoine (Interscience, New York, 1964), Sec. 5.3] have considered the same problem from a somewhat different point of view. They considered the simple quartic Ising lattice and introduced at each vertex point a sublattice to generate what is equivalent to our vertex weights. It can be shown that the free-fermion condition (15) is always an identity in their considerations provided that the sublattice introduced at each vertex is planar. The condition (15) therefore also reflects the solubility of planar Ising lattices.

[17]Green and Hurst (Ref. 15) have evaluated some derivatives of the partition function (16). Our discussions are simpler because of the special form of the vertex weights (2) and the use of relation (19).

[18]J. Stephenson, Can J. Phys. 47, 2621 (1969).

[19]J. Stephenson, J. Math. Phys. 11, 420 (1970).

[20]That the present model is identical to the modified F model (MF) is proved in the Appendix of Ref. 7.

[21]See, for example, E. W. Montroll, in *Applied Combinatorial Mathematics*, edited by E. F. Beckenbach (Wiley, New York, 1964), Chap. 4.

[22]The usefulness of this dimer city has also been observed by M. E. Fisher (private communication).

[23]Except with the replacement of b, c by $-b$, $-c$ in (16). But these are equivalent expressions.

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Elastic Properties of MnF_2 †

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The elastic properties of MnF_2 have been investigated in the temperature range $4.2 < T < 330$ K using a cw transmission technique. The measured values at $T = 300$ K of the six adiabatic elastic constants in units of 10^{11} dyn/cm^2 are $c_{11} = 10.24$, $c_{33} = 16.55$, $c_{44} = 3.185$, $c_{66} = 7.208$, $c_{12} = 7.95$, and $c_{13} = 7.07$. The elastic Debye temperature calculated from the low-temperature elastic-constant data is $\Theta_D(E) = 261.6$ K. Using the force-constant model of Matossi for the rutile structure and the published frequencies of the Raman-active vibrational modes, theoretical values of the elastic constants are calculated and found to be in fair agreement with experiment. Each elastic constant [except $c' = \frac{1}{2}(c_{11} - c_{12})$] shows the expected linear decrease with increasing temperature in the region $T \gtrsim \Theta_D(E)$. On the other hand, c' has not yet reached this limiting high-temperature behavior at $T = 330$ K. The effects on the elastic properties of volume magnetostrictive coupling to the spin fluctuations near the Néel temperature ($T \approx T_N$) and of linear magneto-elastic coupling to the magnetic modes in the antiferromagnetic state ($T < T_N$) are discussed. The elastic constant c_{44} is found to have a component at low temperatures ($T \ll T_N$) decreasing approximately as T^{-1}. This striking phenomenon is discussed on the basis of two possible processes, neither of which provides a completely satisfactory explanation.

I. INTRODUCTION

The study of the propagation of ultrasonic waves in antiferromagnetically ordered media has proven to be a valuable tool for the investigation of a variety of phenomena. Velocity measurements are used to measure the elastic[1,2] and magneto-elastic coupling constants,[3,4] attenuation and velocity measurements are used to study dynamic critical phenomena in the neighborhood of the Néel temperature[5]; anomalies in the elastic properties at the spin-flop transition have been used to study the magnetic phase diagram[6]; and finally, it has been shown that in certain systems resonant nuclear spin-phonon interactions can be studied.[7,8]

In this paper, a systematic study of the elastic properties of MnF_2 is reported. The adiabatic elastic constants associated with longitudinal and transverse ultrasonic propagation along each of the symmetry directions [001], [110], and [100], as well as the nonsymmetry direction perpendicular to the (011) plane have been measured in the temperature region from 4.2 to 330 K. These measurements provide a complete description of the elastic properties of this material.

MnF_2 is an attractive material in which to study