

$$\eta^{\pm} = [p_x \pm i(m\omega x + \hbar k_y)] / (2m\hbar\omega)^{1/2}, X_{\mu} \text{ are given by}$$

$$EX_{\mu} = [\epsilon_{\mu} + \hbar^2 k_z^2 / 2m + (\eta^{-}\eta^{+} - \frac{1}{2})\hbar\omega]X_{\mu}$$

$$+ \sum_{\mu'} [\hbar k_z v_{\mu\mu'} + \epsilon_{\mu\mu'} + (2m\hbar\omega)^{1/2}(v_{\mu\mu'}\eta^{+} + v_{\mu\mu'}\eta^{-})]X_{\mu'}. \quad (\text{A6})$$

For substances and a set of bands characterized by the selection rules (3.3) it is easily seen from the well known

properties of the harmonic oscillator eigenfunctions  $\Phi_n(x)$  that

$$X_{\mu}(x) = C_{\mu}(n)\Phi_{n-m(\mu)}(x), \quad (\text{A7})$$

where  $n-m(\mu)=0, 1, 2, \dots$ , and the coefficients  $C_{\mu}$  and energy eigenvalues  $E$  are given by (3.4) and (3.5). Equations (A7), (A5), and (A1) prove Eqs. (3.1) and (3.4).

## Critical Concentrations in Magnetism

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In the application of the Bethe-Peierls-Weiss method to dilute magnetism, Smart has exaggerated the interaction between a cluster and its surroundings by retaining the uniform molecular field. The calculation is improved by allowing the molecular field to vary from site to site on the first shell of the cluster, thereby taking into account the fact that this interaction is sensitive to the relative location of magnetic atoms in the first shell of the cluster and in the surrounding layer. For the three-dimensional lattices treated, the critical concentration is very nearly  $2/z$ , where  $z$  is the coordination number of the lattice.

THE theory of the behavior of a magnetic system (nearest-neighbor interactions only) when diluted by the substitution of a nonmagnetic component has been treated by a number of authors and the situation has recently been reviewed by Smart.<sup>1</sup> Of particular interest is the dependence of the ferromagnetic transition temperature (Curie point) on the concentration of the magnetic component  $p$ . Only on the molecular field picture does a finite Curie point exist for all finite values of  $p$ . More realistic models predict the occurrence of a critical concentration  $p_c$  below which ferromagnetism does not occur. The simplest of these is the "average coordination number" method of Sato *et al.*<sup>2</sup> This is a modification of the Bethe-Peierls-Weiss<sup>3</sup> (BPW) scheme in which the "first shell" of the cluster is assumed to have just  $pZ$  magnetic atoms,  $Z$  being the number of nearest-neighbor sites characteristic of the lattice. The result,  $p_c Z = 2$ , is physically very suggestive, being just the concentration required for the magnetic atoms to make a linear chain in the crystal on the average. However, these authors<sup>2</sup> also point out that any extension of Bethe's method to the treatment of dilute ferromagnets must take into account all the possible concentration fluctuations which can occur in the system. If nearest-neighbor concentration fluctuations are permitted, then  $p_c = (Z-1)^{-1}$ . This result was obtained by Sato *et al.*<sup>2</sup> using the Takagi-Kikuchi<sup>4</sup> method and the Ising model, and also by Smart<sup>1</sup> in a direct extension

of the BPW method using the classical spin approximation to the Heisenberg model of Brown and Luttinger.<sup>5</sup> In the latter calculation all possible kinds of clusters (magnetic atom populations) were considered with a uniform molecular field describing the interaction of "first shell" magnetic atoms with the remainder of the crystal.

The present work is an extension of Smart's treatment, motivated by the suspicion that the physical problem was inadequately represented because of the neglect of some of the longer range concentration fluctuations. To paraphrase Bethe,<sup>6</sup> the "force" tending to produce order at a given cluster will depend on the *occupation* of the sites in the immediate neighborhood of the cluster under consideration. In particular, with nearest-neighbor interactions alone, the uniform molecular field exaggerates the interaction between a cluster and its surroundings; since this is sensitive to the relative location of the magnetic atoms in the "first shell" of the cluster, and the surrounding layer or "second shell." Indeed, we find that the incorporation of fluctuations in the molecular field results in calculated values for  $p_c$  considerably larger than  $(Z-1)^{-1}$  and quite close to those of Elliott *et al.*<sup>7</sup> and of Frisch *et al.*<sup>8</sup>

We assume that a "first shell" magnetic atom  $i$  is subject to a molecular field proportional to the number of magnetic neighbors  $n_i$  that it has outside the cluster (only lattices with no interactions within the first shell

<sup>1</sup> J. S. Smart, *J. Phys. Chem. Solids* **16**, 169 (1960).

<sup>2</sup> H. Sato, A. Arrott, and R. Kikuchi, *J. Phys. Chem. Solids* **10**, 19 (1959).

<sup>3</sup> P. R. Weiss, *Phys. Rev.* **74**, 1493 (1948).

<sup>4</sup> Y. Takagi, *Proc. Phys.-Math. Soc. Japan* **23**, 44 (1941); R. Kikuchi, *Phys. Rev.* **81**, 988 (1951).

<sup>5</sup> H. A. Brown and J. Luttinger, *Phys. Rev.* **100**, 685 (1955).

<sup>6</sup> H. A. Bethe, *Proc. Roy. Soc. (London)* **A150**, 552 (1935).

<sup>7</sup> R. J. Elliott, B. R. Heap, D. J. Morgan, and G. S. Rushbrooke, *Phys. Rev. Letters* **5**, 366 (1960).

<sup>8</sup> H. L. Frisch, E. Sonnenblick, V. A. Vyssotsky, and J. M. Hammersley, *Phys. Rev.* **124**, 1021 (1961).

TABLE I. Comparison of results for  $p_c$ , the critical concentration required for ferromagnetism.

Lattice, $Z$	$2/Z$	$(Z-1)^{-1}$	Present work	Frisch <i>et al.</i> <sup>a</sup>	Elliott <i>et al.</i> <sup>b</sup>
plane square, 4	0.50	0.33	0.57	$0.581 \pm 0.015$	0.48
simple cubic, 6	0.33	0.20	0.33	$0.325 \pm 0.023$	0.28
body-centered cubic, 8	0.25	0.14	0.24	...	0.22

<sup>a</sup> See reference 8.<sup>b</sup> See reference 7.

are treated). Thus, the molecular field acting on magnetic atom  $i$  is taken to be  $n_i H_1$ . By direct extension of the Smart<sup>1</sup> calculation, and in his notation:

$$Z_P^{(k)}(y, \lambda_0, \lambda_1) = [\alpha(y)]^k \left\{ 1 + \frac{S(S+1)}{6} \right. \\ \times \left[ \lambda_0^2 + 2\lambda_0\lambda_1 \mathcal{E}(y) \sum_{i=1}^k \langle n_i \rangle \right. \\ \left. \left. + \lambda_1^2 \left( \sum_{i=1}^k \langle n_i^2 \rangle + \mathcal{E}^2(y) \sum_{i \neq j=1}^k \langle n_i n_j \rangle \right) \right] \right\}, \quad (1)$$

and

$$Z_Q^{(k)}(y, \lambda_0, \lambda_1) = \left\{ 1 + \frac{S(S+1)}{6} \lambda_1^2 \sum_{i=1}^k \langle n_i^2 \rangle \right\}, \quad (2)$$

where the  $\langle \rangle$  denotes an average over clusters of the given  $k$  value. These expressions for the cluster partition functions replace Eqs. (9) and (10) of reference 1.<sup>9</sup> The average "first shell" magnetic moment is determined by differentiation of the free energy with respect to the average molecular field,  $\langle n_i \rangle H_1$ , and the critical temperature is then defined by the same quadratic equation in  $L(y)$  for both positive and negative  $J$ . Letting  $L(y) = 1$  results in an equation for  $p_c$  whose solution for cases of interest is given in Table I. Our results for the variation of the critical temperature and paramagnetic Curie point with  $p$  will be published elsewhere.

<sup>9</sup> In Eq. (10) of reference 1, the factor  $[\alpha(y)]^k$  should not appear.

Our values of  $p_c$  may appropriately be compared with critical percolation probabilities for the site problem. These have recently been estimated by a Monte Carlo procedure by Frisch *et al.*<sup>8</sup> and, as shown in Table I, our values fall within their limits of accuracy.

By considering the paramagnetic susceptibility of a dilute ferromagnet at  $T=0^\circ\text{K}$  as a function of  $p$ , Elliott *et al.*<sup>7</sup> found values of  $p_c$  which lie fairly close to  $2/Z$ . As Table I shows, the present results are reasonably close to, but somewhat larger, than theirs. For purposes of comparison we have evaluated the susceptibility at absolute zero in the present scheme and find the singularity appropriately occurring only at  $p = p_c$ . However, the expansion of our result as a power series in  $p$  has a radius of convergence smaller than  $p_c$ . (There is a pole on the negative real axis.) While it is not clear that our series is comparable to that of Elliott *et al.*, we feel that their reliance on a limited number of terms may well not be justified by a more thorough analysis.

In this connection we note that Domb and Sykes<sup>10</sup> have considered power series expansions of the mean cluster size in various lattices, which are said to be closely related to the series expansions obtained by Elliott *et al.*<sup>7</sup> More terms are considered and, as a result, all the critical concentrations obtained by Domb and Sykes satisfy the inequalities derived by Fisher,<sup>11</sup> as do ours. However, the value of  $p_c$  which Domb and Sykes obtained for the simple-cubic lattice, the only three-dimensional lattice they treated, is no different from that obtained by Elliott *et al.*

We expect that a calculation of the critical percolation probabilities for the bond problem may be made by the extended BPW method only at the level of Bethe's<sup>6</sup> second approximation. This is being pursued at present.

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<sup>10</sup> C. Domb and M. F. Sykes, Phys. Rev. **122**, 77 (1961).

<sup>11</sup> M. E. Fisher, J. Math. Phys. **2**, 620 (1961).