equations appearing in the papers of BE and GBI.

In summary, we have presented a new method of solution for the BE model. Our method has not only the important advantage of mathematical simplicity, but it also provides a transparent physical picture of the model system. We utilize the equivalence of the BE model to a set of $3N^2$ linear chains of the Mattis-Schultz type. This equivalence exists as a consequence of the very

¹G. A. Baker, Jr. and J. W. Essam, Phys. Rev. Lett. 24, 447 (1970).

²M. Fisher, Phys. Rev. <u>176</u>, 257 (1968).

³L. Gunther, D. J. Bergman, and Y. Imry, Phys. Rev. Lett. 27, 558 (1971).

⁴D. C. Mattis and T. D. Schultz, Phys. Rev. <u>129</u>, 175 (1963); D. C. Mattis, The Theory of Magnetism (Harper and Row, New York, 1965), pp. 239-242.

⁵Rather than explicitly include terms in the Hamiltonian to describe the action of external forces, BE

PHYSICAL REVIEW B

special form of the BE model. In particular, it is essential that the lattice be chosen as simple cubic and that the interatomic shear terms are excluded from consideration. If either of these features are altered, the correspondence no longer holds and the altered model cannot be diagonalized by any available method.

I wish to acknowledge valuable comments made by C. Domb, P. Gluck, and N. Wiser.

evaluate the partition function constraining the surface atoms to their appropriate crystal faces. By contrast, GBI utilize the so-called λ ensemble, which is equivalent to our procedure of utilizing the usual canonical ensemble based on the Hamiltonian of (1). Their parameter λ equals our F.

⁶G. A. Baker, Jr. and J. W. Essam, J. Chem. Phys. $\frac{55}{^{7}}$, 861 (1971). 7 A full discussion of this point is given by M. Luban

and H. Novogrodsky, Phys. Rev. B 6, 1130 (1972).

VOLUME 7, NUMBER 5

1 MARCH 1973

Comment on Mills's "Surface Effects in Magnetic Crystals near the Ordering Temperature"

A. Sukiennicki Institute of Physics, Technical University, Warsaw, Poland

L. Wojtczak

Institute of Physics, University of Ilódź, Ilódź, Poland (Received 30 December 1971)

The Mills result, that the magnetic order at the surface of a semi-infinite crystal appears in a temperature above the Curie point of the infinite crystal, is considered. In the light of this note it is evident that as the Curie point is approached from above the long-range order occurs at one temperature in the whole sample.

Surface effects discussed in one of Mills's recent papers¹ are interesting not only from the point of view of semi-infinite magnetic crystals, but also, and we may even say, first of all, from the point of view of thin-film properties. In the theory of thin films the surface effects and their special role have been known for many years and widely discussed in literature.² The equations-based on the effective-molecular-field methods-for magnetization of thin films were introduced by Valenta³ and developed by Pearson⁴ and Wojtczak.⁵ From Refs. 2-5 it follows that the spontaneous magnetization of the whole sample disappears in one temperature contrary to intuitive suggestions given by Brodkorb.⁶ A similar situation occurs in the case discussed by Sukiennicki⁷ where the spontaneous magnetization of double films coupled by exchange interactions does not vanish locally, but at one temperature corresponding to the Curie temperature of the film with a greater exchange integral.

Since a semi-infinite crystal may be considered as a particular case of a thin film, the results obtained in the thin-film theory refer also to the behavior of spontaneous magnetization at the surface of a semi-infinite crystal. From this point of view we can state that the spontaneous magnetization of a semi-infinite crystal disappears at one temperature. However, Mills reports¹ that from the molecular-field theory it follows that the magnetic order-defined as the average spontaneous magnetization at a lattice site-in a surface layer will appear within the interval of temperatures $T_C < T < T_C^s$ if the exchange integral in a surface layer is sufficiently greater than inside a crystal. T_c^s denotes here a temperature in which the denominator in the formula of the response of the spins in the surface layer to the external field becomes singular and T_c is the Curie temperature of the infinite crystal. A contradiction between the above results inclined us to present this paper.

The molecular-field equations given by Valenta³ for spontaneous magnetization of thin films, after reducing them to the case of semi-infinite crystals with simple cubic lattice and (100) orientation of the surface, are of the form

$$\eta(1) = B_{\mathcal{S}}\left[\frac{S}{2\tau(S+1)} \left(4 \frac{I_s}{I} \eta(1) + \eta(2)\right)\right], \qquad (1)$$

$$\eta(\nu) = B_{S}\left(\frac{S}{2\tau(S+1)} \left[\eta(\nu-1) + 4\eta(\nu) + \eta(\nu+1)\right]\right),$$

 $\nu \in (2, \infty)$

(2)

where

$$\eta(\nu) = \frac{\langle S^{\mathbf{z}}(\nu) \rangle}{S} \tag{3}$$

is an order parameter, $\tau = T/T_c$, and T is the temperature of the crystal; ν labels the monatomic layer parallel to the surface. The exchange integrals I_s for surface atoms are different from interactions I inside the crystal; evidently I > 0, $I_s > 0$. $B_s(x)$ is the Brillouin function and S is the spin value per atom.

In the case of ferromagnets $\eta(\nu)$ is the positive length of the relative spontaneous magnetization, i.e., $\eta(\nu) \ge 0$. Thus, if $\eta(\nu) = 0$ for arbitrarily chosen ν in a given temperature, then it follows from Eq. (2) that $\eta(\nu+1) + \eta(\nu-1) = 0$, that is, $\eta(\nu-1) = 0$ and $\eta(\nu+1) = 0$ which, from Eq. (1), leads to the vanishing of $\eta(1)$ in the same temperature even if $I_s \ne I$. This fact evidently shows that the spontaneous magnetization of the whole semi-infinite crystal disappears at one temperature.

From the physical point of view the above formal result may be interpreted in the following way. As long as the spontaneous magnetization occurs in a certain atomic layer parallel to the surface of a semi-infinite crystal it produces nonvanishing molecular fields in neighboring layers; these fields cause the occurrence of spontaneous magnetizations in the corresponding layers. It seems thus that the existence of spontaneous magnetization in a semi-infinite crystal is self-consistent from the point of view of the molecular-field method.

Thus, there arises the question how, in the light of the above, we should understand Mills's result¹ that the magnetic order of a surface layer appears in temperatures higher than the magnetic order inside a crystal-if $I_s > \frac{5}{4} I$. We believe that this misunderstanding is due to confusing various definitions of magnetic orders used in the paper.¹ In fact, the critical temperature calculated by Mills is a temperature at which the magnetic susceptibility is infinite. On the other hand, this result does not necessarily have to mean that in this temperature there appears magnetic order understood in the sense of the above-introduced definition-formula (3). The example of the phase transition discussed by Stanley and Kaplan² in the case of an isotropic Heisenberg model of a two-dimensional ferromagnet shows evidently that such a situation may occur.

In the light of the above considerations it seems that as the temperature T_c is approached from above, the long-range order occurs at one temperature T'_c equal to the Curie temperature of a semi-infinite crystal (it is evident that T'_c may be higher than T_c). This means that the locality of a temperature of this kind of phase transition does not exist, while it is not impossible that at the temperature $T_c^s > T_c$, where the susceptibility of a surface layer is infinite, there appears a region at the surface, with the magnetic order understood in the sense of Stanley and Kaplan, for which there may exist a local character of its appearance.

ACKNOWLEDGMENT

We wish to express our sincere thanks to Professor S. Szczeniowski for valuable remarks and for the critical reading of the manuscript.

- ²A. Corciovei, IEEE Trans. Magn. 4, 6 (1968).
- ³L. Valenta, Czech. J. Phys. 7, 127 (1957).
- ⁴J. J. Pearson, Phys. Rev. 138, A 213 (1965).
- ⁵L. Wojtczak, J. Phys. (Paris) 30, 578 (1969).

- ⁶W. Brodkorb, Phys. Status Solidi 16, 225 (1966).
- ⁷A. Sukiennicki, Czech. J. Phys. 21, 436 (1971).
- ⁸H. E. Stanley and T. A. Kaplan, Phys. Rev. Lett. **17**, 913 (1966).

¹D. L. Mills, Phys. Rev. B 3, 3887 (1971).