(43). Such experiments are tedious and difficult, however, and have not so far been carried out.

There still remains the unexplained constant contribution to λ, independent of temperature and mean free path. It is difficult to conceive of this as arising from the decay of the spin wave into other elementary excitations, since such a process would surely show a temperature dependence[8,9] over the temperature range 100–600 °K. Therefore, we feel it likely that this source of damping is some sort of inhomogeneous line broadening, perhaps arising from a surface phenomenon. No detailed mechanism for this constant contribution has been found to date, however.

In summary, we have concluded that, by virtue of the spin-orbit interaction, a spin wave can decay directly into a particle-hole pair, the decay not conserving spin. This particular source of damping has an anomalous character and depends on detailed properties of the Fermi surface.

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PHYSICAL REVIEW B VOLUME 6, NUMBER 7 1 OCTOBER 1972

Evidence for Fisher's Droplet Model in Simulated Two-Dimensional Cluster Distributions

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The distribution function n_l of clusters with l reversed spins has been calculated for two square $N \times N$ Ising lattices ($N = 55$ and 110) at various temperatures $T < T_c$ using periodic boundary conditions. These calculations strongly support the cluster model proposed by Fisher and yield the exponent $\tau = 2.1 \pm 0.1$. However, for small l, T not close to T_c, and $H \neq 0$, we find considerable deviations.

Fisher's cluster model[1–3] is a semiphenomenological description of critical phenomena in Ising spin systems, leading to a physical interpretation of the static scaling relations.[3,4] Within the framework of the lattice-gas terminology it can also be applied to the gas-liquid phase transition. These results agree surprisingly well with experiment for gases[5–7] and have also been used in the nucleation problem[8–10] to calculate the formation rate of large droplets in a supersaturated vapor. Perhaps this model can also contribute to a better understanding of the phenomena near tricritical points.[11–13] It should be noted, however, that it fails above the critical temperature T_c.[6]

Attempts to remove this deficiency[14] are rather speculative, including the proposed correction terms[15,16] to the original model,[1-3] which are necessary to get corrections to the scaling equation of state.[16,17]

In view of this situation, it appeared desirable to examine the range of validity of the model more closely. We hope to clarify this point by extending earlier Monte Carlo simulations of critical properties and metastable states to a two-dimensional Ising model.[18] In doing so, we calculated the distribution function of clusters with l reversed spins in a two-dimensional Ising model with periodic boundary conditions by means of the Monte Carlo technique.

In the first part of our paper we summarize the essential predictions of Fisher's cluster model. In the later parts we present our results and discuss the range of validity of this model.

Using the magnetic notation, the cluster model[1-3] leads to the following expression for the free energy per spin:

$$F = -\mu H - U_0 - k_B T \sum_{l=1}^{\infty} n_l . \tag{1}$$

U_0 is the interaction energy in the fully aligned state, H is the magnetic field, μ is the magnetic moment per spin, and n_l is the cluster distribution function varying as[1-3]

$$n_l = q_0 \exp\left\{-\left[a\left(\frac{T_c - T}{T_c}\right)\frac{J}{k_B T}\, l^{\sigma} + \frac{2\mu H}{k_B T}\, l\right]\right\} l^{-\tau} \tag{2}$$

as $l \to \infty$ with $T \simeq T_c$. Note that two exponents, τ and σ, and two constants, q_0 and a, are introduced in an *ad hoc* fashion in the model. The critical exponents[3] can then be expressed in terms of τ and σ, e.g.,

$$\beta = (\tau - 2)/\sigma, \quad \delta = 1/(\tau - 2) . \tag{3}$$

Now σ can be related (roughly[14]) to the variation of the mean cluster surface S_l with cluster size l, $S_l \propto l^{\sigma}$. It is plausible that σ should be close to the geometrical estimate $\sigma_g = (d-1)/d$, where d is the dimensionality of the system. From the exact solution of the $d=2$ Ising model[19,20] one derives[1-3] $\sigma = \frac{8}{15}$. This value has been confirmed by a Monte Carlo study of surface areas of clusters,[21] taking thermal fluctuations into account. Given this value of σ and invoking the scaling relations, one finds $\tau = \frac{31}{15}$.[3,19,20] In view of this, a test of the droplet model amounts to calculating τ. If it turns out that the model fails, recent physical applications[5-16] would appear to be data-fitting procedures. On the other hand, if it turns out to be correct, the applications mentioned[5-16] are justified *a posteriori*, and the range of validity of the model (and necessary corrections) can be specified. Furthermore, one would have a reasonable model for all kinds of nucleation and clustering processes in anisotropic magnets, binary systems such as alloys, etc., and thus a wide class of physical problems could be described on this basis.

For our $N \times N$ Ising system with periodic boundary conditions we have the Hamiltonian

$$\mathcal{H} = -\sum_{i,j} J_{ij}\sigma_i\sigma_j - HN^2 m, \quad \sigma_i = \pm 1 \tag{4}$$

$$J_{ij} = \begin{cases} J & \text{if } i \text{ and } j \text{ are nearest neighbors} \\ 0 & \text{otherwise,} \end{cases} \tag{5}$$

$$m = (\mu/N^2)\textstyle\sum_i \sigma_i .$$

In the Monte Carlo technique[22,23] one generates a Markovian chain of configurations of the spin system using the transition probabilities

$$W_i(\sigma_1 \cdots \sigma_{N^2}) \propto \tfrac{1}{2}[1 - \sigma_i \tanh((H\mu + \textstyle\sum_j J_{ij}\sigma_j)/k_B T)] . \tag{6}$$

We generated about 2×10^4 to 16×10^4 Monte Carlo[24] steps per spin for one set of variables (N, T, $H=0$), which guaranteed a sufficient statistical accuracy and we simulated systems with $N \times N = 55 \times 55$ and 110×110. At every Monte Carlo step per spin (k) the cluster distribution n_l^k is evaluated by counting[25] all the clusters. A "cluster" is defined as a group of reversed spins linked together by (at least one) nearest-neighbor bond. It turns out that distributions n_l^k at consecutive steps k are almost statistically independent, and we can derive $n_l \equiv \langle n_l^k \rangle$ from $l=1$ to $l=100$ with good statistical accuracy. The temperatures used are $J/k_B T = 0.448$, 0.459, and 0.449; since $J/k_B T_c = \frac{1}{2}\ln(\sqrt{2}+1)$ in the infinite square lattice[19] these values correspond to $|\epsilon| = |(T - T_c)/T_c| = 0.016$, 0.040, and 0.117, respectively. From the "raw data" for n_l (compare Ref. 18) it is immediately clear that Eq. (2) can be used as a rough description. Since Eq. (2) is expected to hold for large l only, we omit values of n_l from $l=1$ to $l=10$ from the first stage of the data evaluation procedure. Using Eq. (2) in a fitting program for $10 \leq l \leq 100$, we find

$$\tau = 2.1 \pm 0.1 , \tag{7}$$

which is in very good agreement with the predicted value $\tau = \frac{31}{15}$. We get the same good fit of τ for a broad range of values a and σ with $0 < \sigma < 1$; since the value of σ has been confirmed in a previous paper[21] it is advantageous to take for σ and τ the theoretical values. To demonstrate the range of validity of Eq. (2) we plot $\log_{10} n_l l^{\tau}$ vs l in Fig. 1. For small l ($l \leq 7$) one sees some deviations from the asymptotic predictions. Thus $n_l l^{\tau}$ is essentially constant in this region and there is no indication of an exponential factor. For large l the data fit nicely to the theoretical curves, establishing the

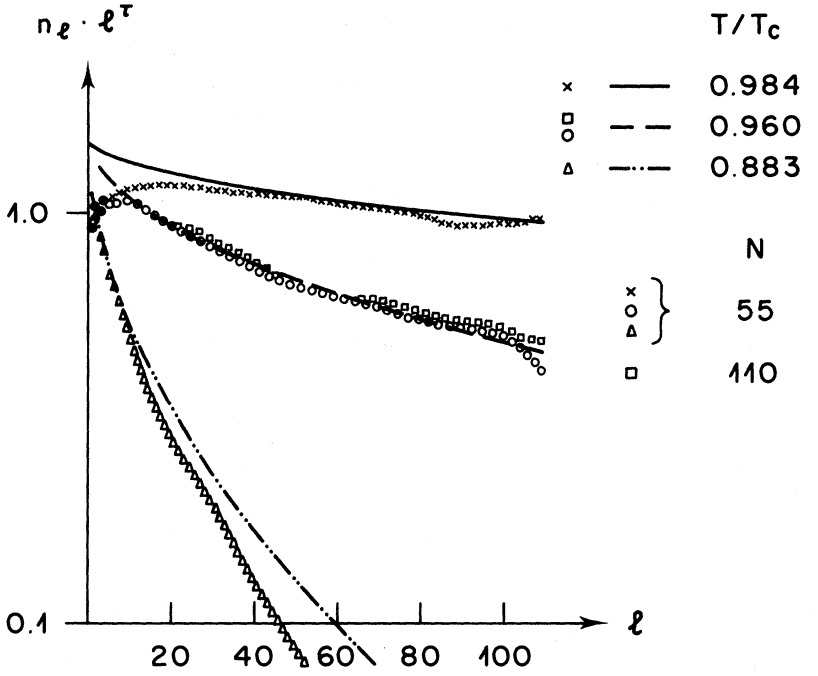

Fig. 1. Cluster distribution ($n_l l^\tau$) for three temperatures and two lattices in units of n_1^g. The curves represent Fisher's droplet model ($\tau = \frac{31}{15}$, $\sigma = \frac{8}{15}$). Circles, crosses, etc., are the Monte Carlo results.

occurrence of an exponential. The slope is indeed roughly proportional to $|\epsilon| = |(T - T_c)/T_c|$ as expected. The behavior for extremely small l ($l = 1, 2, 3$) can be understood in terms of geometrical considerations (n_l^g), by explicitly considering the possibilities of forming clusters of l spins at the square lattice and considering their energies. Excluded volume corrections are made only to "first order."[26,27] In Ref. 16 it was suggested to add a correction term $-b(J/k_BT)\, l^{\sigma-1}$ to the argument of the exponential function in Eq. (2). However, our results are rather insensitive to the argument of an exponential correction term. In any case, a term of this type cannot account for the even-odd oscillations[27,28] at small l which are seen in Fig. 1.

Finally, we have performed similar calculations for systems with free boundaries instead of periodic ones, and in some cases also for $H \neq 0$. In neither case, was a similarly good agreement with the droplet model obtained. Taking into account free surfaces, which have pronounced effects on critical phenomena,[29] such discrepancies must be expected. However the disagreement observed for $H \neq 0$ might indicate some more serious limitations of the droplet model.

To summarize, we succeeded in calculating the cluster distribution n_l of the two-dimensional Ising model and the exponent τ which describes the decay of this function at the critical point. We found good agreement with the value which explains the critical exponents of the Ising model in the framework of the droplet picture. The range of validity $T \approx T_c$ ($H = 0$) is found to be $l \gtrsim 7$. The deviations for smaller l are explained by geometrical arguments. In some of the physical applications[5-7] the model has been used down to $l = 1$. Our results indicate that such extrapolations are doubtful. However, since the coordination numbers of a three-dimensional system are larger, the geometrical effects for small l are expected to be less important in that case. Our further investigations will include the study of a three-dimensional Ising model, and the comparison of the recently derived lifetimes of metastable states[18] with predictions based on the cluster model.

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[25]At every row one counts the reversed spins and stores their positions. Reversed spins in the same row which are nearest neighbors belong to the same cluster, of course. In the next row one has to add those reversed spins which are nearest neighbors of reversed spins of the preceding row to the appropriate previous clusters, etc.
[26]Given the magnetization $\langle m \rangle$ at some temperature, a cluster with l spins can be distributed only over $N(1-\langle m \rangle) - l$ sites on the average. We do not consider the "higher-order" excluded volume which is due to the fact that no spin of the l cluster may be a nearest neighbor of a spin belonging to any other cluster existing in the system. A correct treatment of these effects would lead to the correct low-temperature expansion of the n_l, see Ref. 27.
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PHYSICAL REVIEW B VOLUME 6, NUMBER 7 1 OCTOBER 1972

Heterogeneous Nucleation and Fisher's Droplet Picture

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Corrections to the classical theory of heterogeneous nucleation on ions and small "dust" particles are calculated for water with Fisher's droplet picture. Our results do not differ greatly from Fletcher's and other classical theories; but our derivations should give a more unified and reliable description of the preexponential factors in the nucleation rate. If the vapor contains small dust particles (~10 Å) and ions, then at low temperatures the nucleation will occur mainly on small particles, at intermediate temperatures on ions, and near the critical point homogeneous nucleation will dominate. We suggest measuring such temperature-dependent effects.

This work applies our previous homogeneous nucleation theory[1] to the heterogeneous gas-to-liquid nucleation on ions and small (~10 Å) particles, for temperatures between the triple point and the critical point. Heterogeneous nucleation theories[2,3] of this kind are based on homogeneous nucleation theories,[1,4] and at present[5] there is no general agreement about the correct form of homogeneous nucleation theory. The controversial problems[5] of rotational, translational, configurational, and replacement partition functions still appear to be unclear. Also, most of the studies on homogeneous nucleation have been restricted to the temperature near the triple point. It is our suggestion[1] that a study of the temperature dependence in the whole region between the triple point and the critical point should give more information about the approximation underlying the nucleation theory, e.g., the capillarity approximation, bulk surface tension vs microscopic surface tension.

In Ref. 1 we proposed a theory for homogeneous nucleation for this temperature region which also circumvented the controversial problems of the rotation-translation "paradox" by extrapolating Fisher's droplet picture[6] from the critical point