Violation of Dynamical Scaling for Randomly Dilute Ising Ferromagnets near Percolation Threshold

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Strongly anisotropic magnets at low temperatures with many (quenched) nonmagnetic impurities are shown to deviate from usual dynamical scaling assumptions near the critical concentration; instead of the time, the logarithm of the time is the basic variable. Analysis of previous Monte Carlo data confirms the static droplet picture used here, which fulfills for the first time the homogeneity, analyticity, and symmetry requirements of static scaling.

If in a spin-$\frac{1}{2}$ Ising model with nearest-neighbor ferromagnetic interactions a fraction $1-p$ of spins is replaced randomly by nonmagnetic atoms (zero exchange energy), then the Curie temperature $T_C(p)$ decreases from $T=T_C(1)$ at $p=1$ to $T_C=0$ at $p=p_c$ (percolation threshold). For $T \ll T_C(1)$, all spins connected by interactions have to be parallel. Thus a "cluster," defined as a set of interaction-connected spins, acts like a single large ("superparamagnetic") spin. For zero magnetic field, the magnetization contributions of the many finite clusters cancel because of their random orientations; spontaneous magnetization exists only for $p>p_c$ where one infinite cluster appears; see Kirkpatrick,[1] Essam,[2] and Shante and Kirkpatrick[3] for reviews. The present paper calculates the critical slowing down for $p \to p_c$ of the response to a change in the magnetic field, using a droplet model.[4,5] Throughout the paper I assume $T \ll T_C(1)$ to be constant. Monte Carlo calculations[6,7] will confirm the present droplet picture, in contrast to other choices.[8,9] Quenched substitutional alloys like *Au*Fe allow possible experimental tests for not too low concentrations (above 15% Fe)[10]; contrary to the work of Smith[11] the present paper does not apply this percolation model to spin glasses with long-range oscillating interactions.

Let c_n be the average number per spin of clusters containing n spins each. Every finite cluster size n gives a contribution nc_nM_n to the magnetization M (measured in units of the saturation magnetization), where the orientation factor is $M_n=\tanh(nh)$ in equilibrium, with $h=$(magnetic field)$\times$(magnetic moment)$/k_BT$. The infinite cluster, oriented either up or down, contains the fraction $1-\sum nc_n$ of spins. (Here and later, the abbreviation $\sum$, the summation over n from 1 to ∞, *excludes* the infinite cluster.) Thus $\sum nc_n=1$ for $p<p_c$ and is <1 for $p>p_c$. The cluster numbers c_n are independent of the magnetic field since the impurities are assumed to be quenched. Thus the equation of state is

$$M=M(h,p)=\pm(1-\textstyle\sum nc_n)+\sum nc_nM_n, \qquad M_n=\tanh(nh), \tag{1}$$

and has the desired symmetry which is so difficult to achieve for the droplet model of pure Ising magnets: $M(-h)=-M(h)$. The "impure" droplet picture[5,8,9] for the c_n assumes clusters of the same n to have the same "perimeter." Similar to Fisher,[12] for $p \gtrsim p_c$ and large n, I assume $c_n \propto n^{-\tau}\exp(\epsilon n^{\sigma})$, where $\epsilon \propto p_c-p$. [In contrast, Refs. 8 and 9 assumed $c_n \propto n^{-\tau}\exp(-b'n)$. The choice $b' \propto |\epsilon|^{-1/\sigma}$ in Ref. 9 leads to undesired nonanalyticities in $M(h,p)$ if $p=p_c$ for fixed $h \neq 0$.]

The dominating contributions to the sum in (1) arise from cluster sizes n around a typical droplet size n_ξ defined as $n_\xi \equiv |\epsilon|^{-1/\sigma}$ or $n_\xi \equiv 1/h$ on the coexistence curve and critical isotherm, respectively. For these cluster sizes the argument of the exponential in c_n is of order unity. Thus, at the critical point $h=\epsilon=0$, the size n_ξ (with droplet radius ~coherence length ξ) diverges. The summation in (1) gives an additional factor n_ξ, resulting in a spontaneous magnetization $M \propto \pm|\epsilon|^{\beta} = \pm n_\xi^{\tau-2}$. In this way the assumption for the cluster numbers c_n with its two phenomenological exponents σ and τ gives the static-scaling results customary for such simple droplet models[12,5]: Along the coexistence curve, the free energy varies as $(-\epsilon)^{2-\alpha}$, the spontaneous magnetization as $(-\epsilon)^{\beta}$, and the susceptibility as $(-\epsilon)^{-\gamma}$, whereas along the "critical isotherm" $\epsilon=0$, i.e., $p=p_c$, one has a nonzero magnetic field $h \propto M^{\delta}$. Thus the usual exponents α, β, γ, and δ are defined and related by $2-\alpha=\gamma+2\beta=\beta(\delta+1)$ whereas the droplet exponents are expressed as $\sigma=1/\beta\delta$ and

$\tau = 2 + 1/\delta$, as usual. For the paramagnetic region $p < p_c$, $\epsilon > 0$, the total number of spins in finite clusters must equal the total number of spins, i.e.

$$\sum n c_n(p < p_c) = 1 = \sum n c_n(p = p_c). \quad (2)$$

For this purpose, one can use analogously to Reatto and Rastelli[13]

$$c_n \propto n^{-\tau} e^{\epsilon n^{\sigma}} / (1 + B e^{b \epsilon n^{\sigma}}) \quad (3)$$

with $B = B(b)$ such that Eq. (2) is fulfilled.[13] {More generally,[4] $c_n \propto n^{-\tau} f(x)$, $x \equiv \epsilon n^{\sigma}$, $f(x)$ analytic and universal, $\int_0^{\infty} x^{-1-\beta}[f(\pm x) - f(0)]dx = 0$ for + and < 0 for $-$.} The resulting Eq. (1) fulfills the homogeneity, analyticity, symmetry, and universality requirements of static scaling,[2] the first explicit expression with these properties for general exponents. [Equations (1) and (3) give an essential singularity[9,12] at $h = 0$ as function of h for both $\epsilon > 0$ and $\epsilon < 0$.]

Are these assumptions, which contradict those of Refs. 8 and 9, correct? The Monte Carlo data of Refs. 6 and 7, usually overlooked, test these assumptions, particularly[6] for dimensionality $d = 2$. Although fewer Monte Carlo steps were used than in the cluster simulation for pure Ising systems,[14] the present analysis allows for a more complete test than in Ref. 14 and leads to the following results. (i) For $p < p_c$, $n^{\tau} c_n$ first increases with n, then decreases, as expected from Eq. (3). (ii) For fixed n, the cluster numbers $c_n(p)$ have maxima at $p_{max} < p_c$, as Fig. 1 shows: $p_c - p_{max} \propto n^{-\sigma} = n^{-0.36 \pm 0.1}$. (iii) At $p = p_c$, an inset in Fig. 1 shows $c_n \propto n^{-\tau} = n^{-2.0 \pm 0.1}$ for two dimensions ($n^{-2.2 \pm 0.2}$ for $d = 3$). (iv) Figure 2 indicates that for fixed $p = 0.55 > 0.50 = p_c$, the cluster numbers c_n decay as $\exp(-\text{const} \times n^{\sigma})$ for large n, again with $\sigma = 0.36$. (v) The inset in Fig. 2 suggests analytical behavior for fixed cluster sizes: $c_n/c_{n'} \propto 1 - \text{const}(p - p_c) + \ldots$ at a fixed ratio n/n', as required by Eq. (3). The alternative theories[8,9] with $c_n \propto n^{-\tau} e^{b'n}$ predict for $\sigma \neq 1$ $p_{max} = p_c$ in Fig. 1, a line with slope 1 in Fig. 2, and a maximum at $p = p_c$ for the curve in the inset of Fig. 2; thus whereas the present theory[5] is consistent with the Monte Carlo data,[6] the previous theories[8,9]

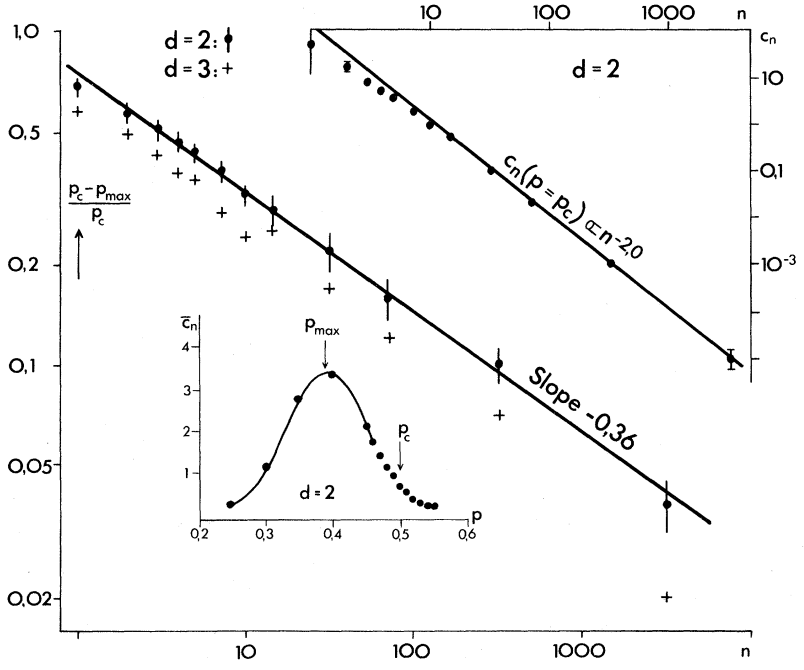

FIG. 1. Log-log plot of the Monte Carlo values for p_{max}, where the cluster numbers $c_n(p)$ reach their maximum at fixed size n as a function of concentration p, versus n. The inset at the center shows a typical curve $\bar{c}_n$ ($= \sum_n c_n$, $n = 21$ to 50, in units of $10^{-3} \times$number of lattice sites) for the triangular lattice. The present theory predicts $p_{max} - p_c \propto n^{-\sigma} = n^{-1/\beta\delta}$, whereas $p_{max} = p_c$ in the theories of Refs. 8 and 9. The inset in the upper right-hand corner is a log-log plot of the "critical cluster numbers" $c_n(p = p_c) \propto n^{-\tau}$ versus n, normalized to $c_{10} = 1$ and averaged over five two-dimensional lattices. The Monte Carlo raw data are taken from Ref. 6 where, for $n > 10$, only averages of cluster numbers c_n are given for the intervals of n between 11 and 20, between 21 and 50, between 51 and 100, between 101 and 1000, and between 1001 and 10 000 (reduction of statistical errors!). The n in the figures is the geometric mean of the two endpoints of the interval.

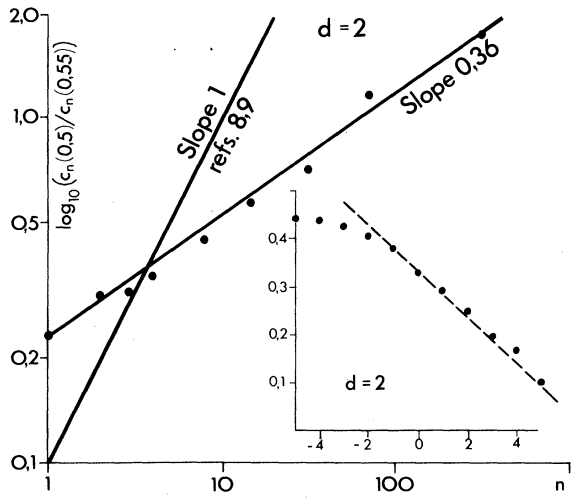

FIG. 2. Log-log plot of $\log_{10}[c_n(p_c)/c_n(p)]$ versus n, where c_n is averaged over triangular-site and square-bond percolation at $p=0.55$; here $p_c=0.50$ exactly. The present theory predicts $c_n(p_c)/c_n(p)=\exp(|\epsilon|n^{\sigma})$ and thus a line with slope σ in this log(log)-log plot, while Refs. 8 and 9 predict $\exp(b'n)$ for this ratio and thus a slope of unity. The inset shows as a function of $p-p_c$ (in percent) the ratio

$$\sum_{n=51}^{100} c_n / \sum_{n=21}^{50} c_n,$$

averaged over five two-dimensional lattices. The present theory predicts for this ratio a linear variation around $p=p_c$ whereas Refs. 8 and 9 predicted a maximum at $p=p_c$. The Monte Carlo raw data are taken from Ref. 6.

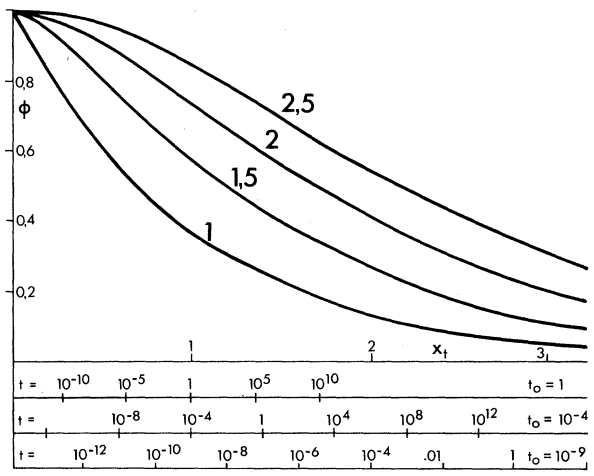

FIG. 3. Predicted ferromagnetic relaxation function $\Phi \equiv \Delta M(t)/\Delta M(0)$, Eq. (4), showing the logarithmic dynamical scaling through the "time" x_t which varies as some power of $\ln(R_0 t)$, as defined after Eq. (4). The three lower scales indicate the real time t (in seconds; 3 yr $=10^8$ sec) for $r=\sigma$, $R_0=10^{14}$ sec^{-1}, and $\ln(R_0 t_0)$ $=\ln(10^{14}t_0)=w/\epsilon \propto 1/(p-p_c)$ equal to 32, 23, and 11.5, respectively. Obviously, true critical behavior will be difficult to observe. (The numbers on the curves give the susceptibility exponent γ.)

are not. In conclusion, the percolation clusters follow a size distribution similar to the one of pure magnets.[14]

[Log-log plots of the paramagnetic susceptibility and ferromagnetic spontaneous magnetization give[1,6,7] $\beta_{d=2} \gtrsim 0.1$, $\gamma_{d=2}=2.23\pm0.2$, $\beta_{d=3}=0.35\pm0.05$, and $\gamma_{d=3}=1.85\pm0.2$, in rough agreement with the series estimates[2] $\beta_{d=2}=0.14$, $\gamma_{d=2}=2.4$, $\gamma_{d=3}=1.7$, and the present direct estimates for σ and τ. Separately[15] I discuss why these exponents differ from the "pure" Ising exponents near $T_C(1)$. Also the static correlation function[13] and the crossover from $T_C(p)>0$ to $T_C(p_c)=0$ are calculated there.[15]]

Now I discuss the dynamic behavior on the ferromagnetic side (p slightly larger than p_c) where the B correction in Eq. (3) is neglected. The clusters change their orientation with a rate R_n; these rates cause the cluster orientation factor $M_n(t)$ in Eq. (1) to lag behind a sudden change in the magnetic field whereas the cluster numbers c_n remain unchanged: $M_n(t)-M_n(t=\infty)\propto\exp(-R_n t)$. Presumably $R_n=R_0\exp(-wn^r)$ due to thermal jumping or quantum-mechanical tunneling over the energy barrier of (Ising) cluster reorientation; e.g., $r=1$, $w\sim1$ for tunneling.[16] If at time $t=0$ a small field h is switched off, then with $M_n(t)=\tanh(nh)\exp(-R_n t)$ we find $M(t)$ from Eq. (1) for $t\to\infty$, $\epsilon\to0$:

$$\Phi(t)\equiv\frac{M(t)-M(\infty)}{M(0)-M(\infty)}=\frac{\int_{x_t}^{\infty}x^{\gamma-1}e^{-x}\,dx}{\int_0^{\infty}x^{\gamma-1}e^{-x}\,dx}\equiv\tilde{\Phi}(x_t), \quad (4)$$

as plotted in Fig. 3. Here $x_t\equiv|\epsilon|[(1/w)\ln(R_0t)]^{\sigma/r}=[\ln(R_0t)/\ln(R_0t_0)]^{\sigma/r}$, which defines t_0 as a decay time. This dependence on $\ln(R_0t)/\ln(R_0t_0)$ is *not* the usual type of dynamical scaling where Φ is a function of the ratio t/t_0 only. Instead a new type of "logarithmic dynamical scaling" is found with $\Phi=\Phi(t'/t_0')$ where $t'\equiv\ln(R_0t)$ and $t_0'\equiv\ln(R_0t_0)$ replace the true times t and t_0.

As the time scales (in seconds) of Fig. 3 indicate, the true equilibrium and the logarithmic-scaling asymptotic decay towards equilibrium will be difficult to observe for very small p_c-p because of the then very long "critical" relaxation times t_0. It should be possible, however, to measure the drastic increase in the decay time, $t_0\propto\exp(\text{const}|p-p_c|^{-r/\sigma})$, if the phase transition

$p = p_c$ is approached at constant T. Also one could test the present scaling prediction for the static equation of state in random substitutional magnetic alloys, in particular the relation $\delta = 1 + \gamma/\beta$.

Thus a simple calculation led to complicated dynamical behavior, a percolation droplet picture consistent with Monte Carlo results was developed, and experiments were suggested as tests for parts of the present predictions. I thank K. Binder for suggesting parts of this work, and him, J. W. Essam, D. P. Landau, H. Müller-Krumbhaar, D. A. Smith, and G. Toulouse for discussions and information.

[1]S. Kirkpatrick, Rev. Mod. Phys. 45, 574 (1973).
[2]J. W. Essam, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1972), Vol. 2, p. 197.
[3]V. K. Shante and S. Kirkpatrick, Advan. Phys. 20, 235 (1971).
[4]K. Binder, D. Stauffer, and H. Müller-Krumbhaar, Phys. Rev. B 10, 3853 (1974), and unpublished.
[5]D. Stauffer, J. Phys. C: Proc. Phys. Soc., London 8, L172 (1975).
[6]P. Dean and N. F. Bird, National Physical Laboratory, Teddington, England, Report No. MA 61, 1966 (unpublished).
[7]K. J. Duff and V. Canella, in *Amorphous Magnetism*, edited by H. O. Hooper and A. M. de Graaf (Plenum, New York, 1973), p. 207, and in *Magnetism and Magnetic Materials—1972*, edited by C. D. Graham, Jr., and J. J. Rhyne, AIP Conference Proceedings No. 10 (American Institute of Physics, New York, 1973), p. 541.
[8]J. W. Essam and K. M. Gwilym, J. Phys. C: Proc. Phys. Soc., London 4, L228 (1971).
[9]C. Domb, J. Phys. C: Proc. Phys. Soc., London 7, 2677, 4552 (1974).
[10]J. A. Mydosh *et al.*, Phys. Rev. B 10, 2845 (1974); V. Canella and J. A. Mydosh, Phys. Rev. B 6, 4220 (1972).
[11]D. A. Smith, J. Phys. F: Metal Phys. 4, L266 (1974), and unpublished.
[12]M. E. Fisher, Physics (Long Is. City, N.Y.) 3, 255 (1967); C. S. Kiang and D. Stauffer, Z. Phys. 235, 130 (1970).
[13]L. Reatto and E. Rastelli, J. Phys. C: Proc. Phys. Soc., London 5, 2785 (1972).
[14]E. Stoll, K. Binder, and T. Schneider, Phys. Rev. B 6, 2777 (1972); K. Binder and H. Müller-Krumbhaar, Phys. Rev. B 9, 2328 (1974); H. Müller-Krumbhaar, Phys. Lett. 48A, 459 (1974), and 50A, 27 (1974); H. Müller-Krumbhaar and E. Stoll, unpublished.
[15]D. Stauffer, to be published.
[16]I. M. Lifshitz and Yu. Kagan, Zh. Eksp. Teor. Fiz. 62, 385 (1972) [Sov. Phys. JETP 35, 206 (1972)]; D. Stauffer, J. Aerosol Sci. 6, 160(A) (1975).

Orientation of Nucleic Acids in High Magnetic Fields

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The magnetic orientation of native and synthetic nucleic acids has been observed for the first time by measuring the Cotton-Mouton effect in magnetic fields B up to 14 T. Our data give evidence that a partial alignment of the nucleic acids takes place in the plane perpendicular to B and that the diamagnetically anisotropic bases are responsible for this orientation. The method reported here represents a new way to determine the persistence length of flexible polymers.

The magnetically induced orientation of large biological systems like cells with diamagnetic anisotropy has been observed[1-3] in magnetic fields around 1 T, and the orientation of polymer aggregates having a size of a few microns has recently been reported[4] for polystyrene solutions subjected to 1.7 T. The availability of higher magnetic fields makes possible the measurable orientation also of single macromolecules in dilute solutions. We report here the first experiments on the orientation of high-molecular-weight native deoxyribonucleic acid (DNA) and of some synthetic nucleic acids in aqueous solution in high magnetic fields up to 14 T. Using these examples we show that the method described here represents a new and general way to determine the persistence length of flexible polymers.

Let us first consider the case of a *rigid rod* made up of N diamagnetically anisotropic subunits with $\chi_\parallel > \chi_\perp$, $\chi_\parallel$ and $\chi_\perp$ being the absolute susceptibility values parallel and perpendicular to the rod axis, respectively. The magnetic energy re-