Computer modeling of the critical dynamics of dilute magnetic materials

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Critical relaxation of the magnetization in a three-dimensional Ising model with nonmagnetic impurity atoms frozen at the lattice sites is modeled on a computer. A 48^3 system with spin concentrations $p = 1.0, 0.95, 0.8, 0.6$, and 0.4 is studied. The dynamical critical exponent z is determined by the Monte Carlo method combined with the dynamical renormalization group method. The results were: $z(p):z(1.0) = 1.0 = 1.97 \pm 0.8, z(0.95) = 2.19 \pm 0.07$, $z(0.8) = 2.20 \pm 0.08, z(0.6) = 2.58 \pm 0.09$, and $z(0.4) = 2.65 \pm 0.12$. We propose the hypothesis that the critical exponents of three-dimensional dilute magnetic materials exhibit stepped universality.

In this work we check the results of our renormalization-group description of the critical dynamics of dilute magnetic materials[1] by means of computer modeling of the critical relaxation of a disordered three-dimensional Ising model for a wide range of impurity concentrations $c_{\text{imp}} = 0.05, 0.2, 0.4$, and 0.6.

During the last two decades many investigators have been studying how impurities and other structural defects affect the behavior of different systems in the presence of phase transitions. The effect of frozen impurities is especially interesting. These impurities are randomly distributed and their presence is manifested as a disturbance of the local temperature. Investigations have shown[2] that frozen impurities change the properties of magnetic materials, whose heat capacity in the homogeneous state diverges at the critical point with exponent $\alpha > 0$. Only systems whose effective Hamiltonian near the critical point is isomorphic to the Ising model satisfy this criterion.

Renormalization group analysis using the ε-expansion[3,4] has revealed that the critical behavior of the dilute Ising model is characterized by a new set of critical exponents, whose values do not depend on the concentration of point impurities in the region $c_{\text{imp}} \ll 1 - p_c$, where p_c is the spin-percolation threshold. The asymptotic convergence of the ε-expansion series for dilute magnetic materials, however, is even weaker than for homogeneous magnetic materials. The equilibrium and dynamical critical behavior of dilute magnetic materials are studied in Refs. 5, 6 and Ref. 1, respectively, directly for three-dimensional systems. Birgeneau *et al.*[7] confirmed experimentally that the statistical critical exponents for impurity systems are different from their values for homogeneous magnetic materials, and the experimental results agree well with the theory. The critical dynamics of dilute magnetic materials has not been studied experimentally. The questions of whether the critical exponents of impurity systems are universal, i.e., independent of the impurity concentration right up to the percolation threshold, or whether there exists a line of fixed points that determines the critical exponents as continuous functions of the concentration, remain unanswered.

Computer modeling of critical phenomena is now becoming an alternative to an actual physical experiment. In Refs. 8 and 9, which are devoted to the modeling of the dilute Ising model, it was observed that the effective critical exponent β for the magnetization is a continuous function of the impurity concentration, and the concept of universality of the critical exponents is confirmed in Ref. 10 within the uncertainties in the values obtained for the susceptibility exponent γ and the correlation length exponent ν for spin concentrations $p = 0.8$, 0.6, and 0.4.

In the present work the critical dynamics of the three-dimensional Ising model was modeled on a computer by the Monte Carlo method for both the homogeneous case and for spin concentrations $p = 0.95$, 0.8, 0.6, and 0.4. There are grounds for assuming that because of specific conservation laws the influence of frozen impurities will be more pronounced in the critical dynamics than in the case of the equilibrium properties.

The Ising model consists of a system of spins S_i, associated with $N = L^d$ sites of a d-dimensional lattice, where L is the characteristic dimension of the lattice. A spin can assume the values $S_i = \pm 1$. This gives 2^N possible configurations $\{S\}$ with energy

$$E = -J\sum_{i,j} S_i S_j - h\sum_i S_i, \tag{1}$$

where the first sum extends over all nearest-neighbor spin pairs, J characterizes the spin interaction energy, and h is the external field coupled to the spins. We consider a ferromagnetic system with $J > 0$.

Ising-model dynamics is customarily described by the conditional probability function $P_s(t) \equiv P(\{S\}, t)$, which satisfied Glauber's kinetic equation

$$\frac{dP_s}{dt} = -P_s(t)\sum_{s'} W(S \to S') + \sum_{s'} W(S' \to S)P_{s'}(t), \tag{2}$$

where $W(S \to S')$ is the probability of a transition of the system from a microscopic state given by the spin configuration $\{S\}$ to a state with the configuration $\{S'\}$. In order that the Markov process described by Eq. (2) converge to the equilibrium state of a Gibbs ensemble with $P_s = \exp(-E_s/kT)$, the detailed-balancing condition must be satisfied: $W(S \to S')P_s = W(S' \to S)P_{s'}$. This relation does not determine the function W uniquely. The function W is usually chosen in the form of Metropolis's function

$$W(S \to S') = \begin{cases} \exp(-\Delta E_{ss'}/kT) & \text{for} \quad \Delta E_{ss'} > 0 \\ 1 & \text{for} \quad \Delta E_{ss'} \le 0 \end{cases} \tag{3}$$

or Glauber's function

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$$W(S \to S') = \exp(-\Delta E_{ss'}/kT)/[1 + \exp(-\Delta E_{ss'}/kT)]. \quad (4)$$

The relation $\langle A(t)\rangle = \Sigma_s A_s P_s(t)$ determines the dynamical evolution of the quantity A_s by means of the function $P_s(t)$—the solution of Eq. (2).

Metropolis's algorithm, which consists of choosing randomly the spin S_i and flipping the spin with probability determined by the function W in Eq. (3), makes is possible to implement directly the Ising-model dynamics with relaxation of the magnetization $m_s(t) = \Sigma_i^N S_i/N$ to the equilibrium value determined by the thermostat temperature T. The time scale t can be associated with the scale $\{S\}$ of successive configurations, assuming that N system sites are chosen randomly per unit time. One time unit corresponds to one Monte Carlo step per spin. In modeling the critical dynamics the initial state of the system is chosen when all spins are parallel ($m_s = 1$) and the temperature of the system is equal to the critical temperature. The critical temperature T_c for dilute magnetic materials is a function of the impurity concentration c_{imp}, decreasing with increasing c_{imp} and vanishing at the threshold concentration $c_{\text{imp}} = 1 - p_c$. For a cubic lattice of Ising spins $p_c \simeq 0.31$ and $T_c(p)$ are equal to: $T_c(1) \simeq 4.5108$, $T_c(0.95) \simeq 4.2571$, $T_c(0.8) \simeq 3.4959$, $T_c(0.6) \simeq 2.4178$, and $T_c(0.4) \simeq 1.2066$ (Ref. 10) in units of J/k.

We have used here the Monte Carlo method, combined with the dynamical renormalization group method,[11] to determine the dynamical exponent z characterizing the critical increase in the relaxation time of the system $t_c \sim |T - T_c|^{-z\nu}$. For this, the system was partitioned into blocks, where a block b^d of neighboring spins was replaced by a single spin whose direction is determined by the direction of most spins in the block. The redefined spin system forms a new lattice with magnetization m_b. Let the magnetization of the initial lattice relax to some value m_1 over a time t_1, and let the redefined system reach the same value m_1 over the time t_b. Then by using two systems with block sizes b and b' and determining the relaxation times t_b and $t_{b'}$ of the block magnetizations m_b and $m_{b'}$ to the same value m_1, the dynamical exponent z can be determined from the relation

$$t_b/t_{b'} = (b/b')^z$$

or

$$z = \ln(t_b/t_{b'})/\ln(b/b') \quad (5)$$

in the limit of sufficiently large b and $b' \to \infty$.

We applied this algorithm to a homogeneous system and to impurity ones with dimensions 48^3 and with the frozen impurity concentrations presented above (the impurities are empty lattice sites, distributed with probability c_{imp}). The size of the system made it possible to partition it into blocks with dimensions $b = 2$, 3, 4, 6, 8, and 12. The procedure of block partitioning of the initial spin and impurity configurations was implemented on the basis of the criterion of spin connectivity. Thus a b^d-dimensional block was considered to be a spin block and replaced by an effective spin oriented in a direction determined by the direction of most spins in the block if the block contained a spin cluster connecting opposite faces of the block. Otherwise, the block was considered to be an impurity block and replaced by an empty site in the renormalized lattice. A relaxation modeling procedure consisting of 1000 Monte Carlo steps per spin was performed for each system with 20–30 runs with different impurity configurations over which the functions $m_b(t)$ were averaged. Figures 1 a–d show plots of the initial and renormalized magnetizations $m_b(t)$, averaged over impurity configurations, with spin concentrations $p = 1$, 0.95, 0.8, and 0.4, respectively, as functions of time.

The computer modeling of the relaxation properties of a three-dimensional homogeneous Ising model, performed in Ref. 12, showed that near the critical temperature the change in the magnetization is characterized by an effective exponential dependence. Our analysis of the relaxation curves $m_1(t)$ at the critical temperature $T_c(p)$ revealed a power-law dependence $m_1(t) \sim t^{-a}$. The following values were obtained for the exponent $a(p)$ with m_1 ranging from 0.65 to 0.45: $a(1) = 0.246 \pm 0.011$, $a(0.95) = 0.236 \pm 0.020$, $a(0.8) = 0.219 \pm 0.018$, $a(0.6) = 0.178 \pm 0.017$, and $a(0.4) = 0.102 \pm 0.017$. The well-known scaling relation for the magnetization

$$m(h, \tau) = \tau^{\beta}\tilde{m}(h/\tau^{\Delta}), \quad (6)$$

where $\tau = (T_c - T)/T_c$ is the reduced temperature, h is the external magnetic field, and β and Δ are the critical exponents, can be generalized for the time-dependent case in the form

$$m(h, \tau, t) = \tau^{\beta}M(h/\tau^{\Delta}, t/t_c) = \tau^{\beta}M(h/\tau^{\Delta}, t/\tau^{z\nu})$$
$$= t^{-\beta/z\nu}\tilde{M}(ht^{\Delta/z\nu}, \tau t^{1/z\nu}) \quad (7)$$

using the asymptotic time dependence of the relaxation $t_c \sim |\tau|^{-z\nu}$. Hence, for $h = 0$ and $\tau = 0$, the power-law character of the relaxation is reflected in the form of the following asymptotic relation

$$m(t) \sim t^{-\beta/z\nu}. \quad (8)$$

According to theoretical calculations $\beta \simeq 0.325$, $\nu \simeq 0.630$,[13] $z \simeq 2.025$,[1] and $\beta/z\nu \simeq 0.255$ for the homogeneous Ising model and $\beta \simeq 0.349$, $\nu \simeq 0.678$,[6] $z \simeq 2.237$,[1] and $\beta/z\nu \simeq 0.230$ for the dilute Ising model. Comparing these values of $\beta/z\nu$ with the exponents $a(p)$ shows that good agreement is obtained for $p = 1$, 0.95, and 0.8 but not for $p = 0.6$ and 0.4.

In order to determine the values of the exponent z independently it is better to employ the relation (5). However, the power-law character found for the relaxation of the magnetization at the critical temperature enabled us to employ, in contrast to Refs. 11 and 12, a different and, we believe, better-founded procedure for processing the curves for the renormalized magnetizations $m_b(t)$. Thus the $m_b(t)$ curves plotted in a double logarithmic scale were approximated by the straight lines $\log m_b = k_b \log t + n_b$ by the least-squares method in intervals Δm_b corresponding best to a power-law variation of m_b. Next, the coefficients k_b were averaged and an average value k_{av} was determined, after which the parameters n_b of the straight lines $\log m_b = k_{\text{av}} \log t + n_b$ were determined by extending the lines through the point of intersection with $\log m_b = k_b \log t + n_b$ at the center of the intervals Δm_b. As a result, the formula for z becomes

$$z = (n_{b'} - n_b)/[k_{av}]g(b/b')]. \quad (9)$$

Sets of values of the exponent z_b corresponding to dif-

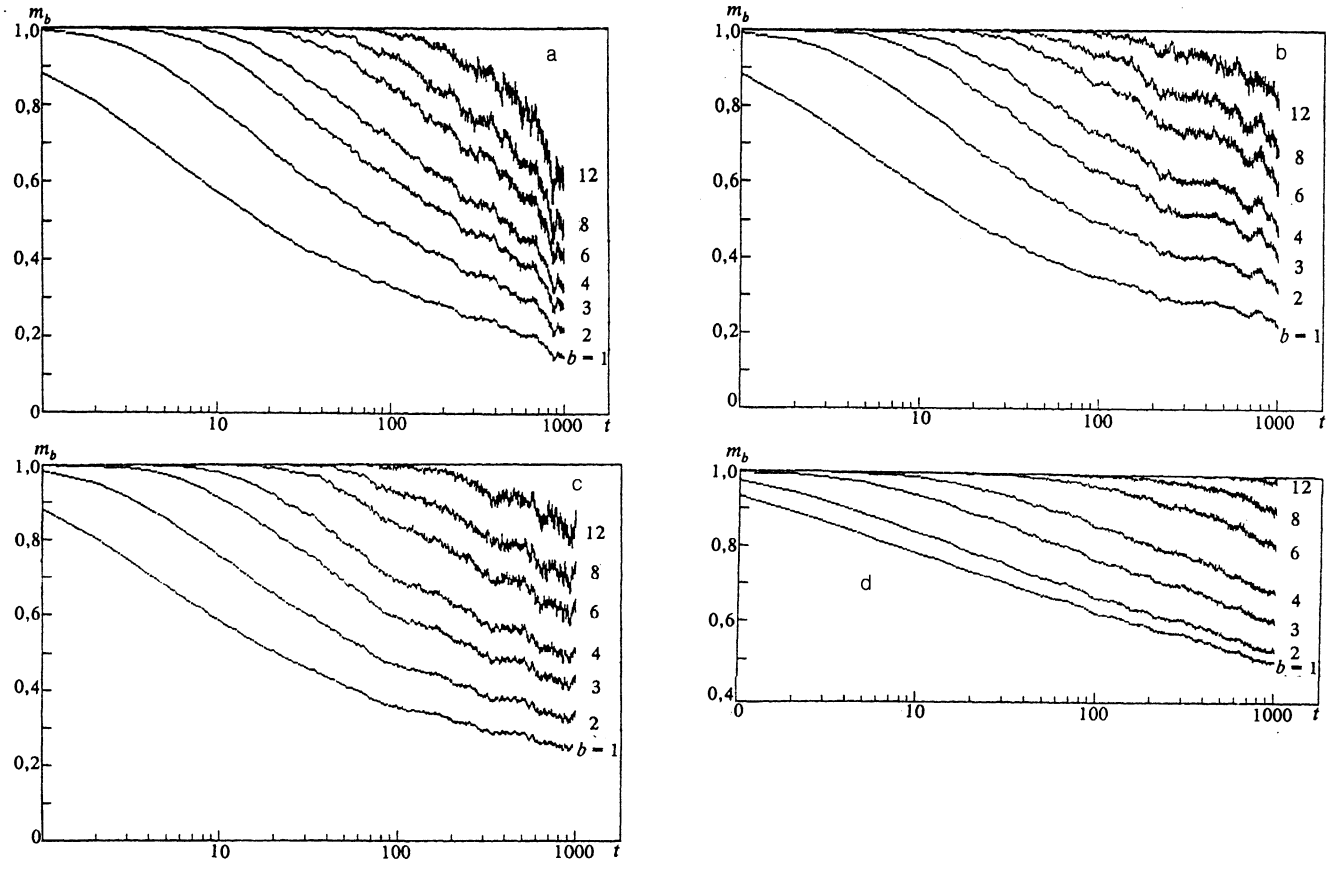

FIG. 1. Initial m_1 and renormalized m_b magnetizations as functions of time for the homogeneous Ising model (a) and for a dilute Ising model with spin concentrations $p = 0.95$ (b), 0.8 (c), and 0.4 (d).

ferent values of b with $b' = 1$ were obtained using the relations (9) (Table I). For impurity systems the renormalization-group-transformation procedure reaches the proven asymptote of m_b as a function of the block-partition parameter b at larger values of b than in the case of a homogeneous system, and for this reason we selected for the analysis the values of the exponent z_b corresponding to $b = 6$, 8, and 12 for impurity systems and $b \geqslant 3$ for homogeneous systems. The obtained dependence of z on b made possible extrapolation to the case $b \to \infty$, assuming that $z_b = z_{b=\infty} + \text{const}\, b^{-1}$. The following results were obtained: for the homogeneous system $z(1) = 1.97 \pm 0.08$ and for the impurity systems $z(0.95) = 2.19 \pm 0.07$, $z(0.8) = 2.20 \pm 0.08$, $z(0.6) = 2.58 \pm 0.09$, and $z(0.4) = 2.65 \pm 0.12$. Hence it is clear that the value of the dynamical exponent for $p = 0.95$ is virtually identical to the value for $p = 0.8$, while for $p = 0.6$ and 0.4 they are equal to within the uncertainties in their values. Taking into consideration the exponent z for a homogeneous system, the values obtained can be divided arbitrarily into three groups differing significantly in magnitude. We note that the value found for the exponent z for a homogeneous system agrees with the value $z = 1.99 \pm 0.03$ obtained in Ref. 12 by Monte Carlo modeling of 128^3-, 265^3-, and 512^3-dimensional systems.

We now compare the computer results with the results obtained by the methods of the theory of crtical phenomena applied to homogeneous and impurity systems. In Ref. 1 we gave a field-theoretical description of the critical dynamics of dilute magnetic materials directly for the three-dimensional case. In the two-loop approximation, using the Padé–Borel summation technique, we obtained the critical exponent $z(p) = 2.237$, valid for impurity concentrations much less than the spin-percolation threshold. A similar calculation performed in the three-loop approximation for the homogeneous Ising system gave the value $z(1) = 2.014$. Comparing the theoretical results with the modeling results

TABLE I. Values of the dynamical exponent z_b obtained using the formula (9) and the extrapolated values $z_b = \infty$ for systems with different spin concentrations p.

b \ p	1,0	0,95	0,8	0,6	0,4
3	2,34 ± 0,07				
4	2,33 ± 0,06				
6	2,21 ± 0,03	2,33 ± 0,04	2,51 ± 0,08	2,45 ± 0,06	2,73 ± 0,06
8	2,12 ± 0,02	2,23 ± 0,02	2,44 ± 0,07	2,53 ± 0,05	2,83 ± 0,05
12	2,04 ± 0,02	2,27 ± 0,04	2,36 ± 0,07	2,51 ± 0,05	2,67 ± 0,04
$z_{b=\infty}$	1,97 ± 0,08	2,19 ± 0,07	2,20 ± 0,07	2,58 ± 0,09	2,65 ± 0,10

shows that they are in good agreement for a homogeneous system and for an impurity system with $p = 0.95$ and 0.8. For $p = 0.6$ and 0.4 the modeling results give a much higher value of the dynamical exponent z. We attribute this to the fact that for a cubic lattice of Ising spins with $p \leqslant p_c^{(\mathrm{imp})} \simeq 0.69$ the impurities form a connecting cluster, which for $T \leqslant T_c$ coexists with a connecting spin cluster right up to the spin-percolation threshold $p_c = 1 - p_c^{(\mathrm{imp})}$. As a result, the spin correlation length in the region $p_c \leqslant p \leqslant p_c^{(\mathrm{imp})}$ is not the only scale determining the behavior of the system near the critical temperature $T_c(p)$. The character of impurity scattering of long-wavelength fluctuations of the magnetization also changes.

By analogy with Refs. 14 and 15 and the works of one of us, Refs. 16 and 17, where the influence of the correlation of the impurities and extended structural defects on the critical properties of disordered systems was investigated, there are grounds for believing that in the region $p_c < p < p_c^{(\mathrm{imp})}$ the existence of an extended impurity structure results in a change in the Harris criterion[2] for the effect of frozen point defects. For this reason, the change in sign of the heat-capacity exponent α (from positive to negative) at a transition from homogeneous to impurity critical behavior in Ising magnetic materials does not limit the new type of critical behavior determined by extended impurity structure.

On the basis of what was said above, we propose the hypothesis of stepped universality of critical exponents for three-dimensional dilute magnetic materials (for two-dimensional magnetic materials such effects do not arise, since $p_c > 0.5$). According to this hypothesis, for dilution $p \geqslant p_c$ five types of different critical behavior can be observed: uniform; impurity type-I for $p_c^{(\mathrm{imp})} < p < 1$ with an influence of point impurities; impurity type-II with $p_c < p < p_c^{(\mathrm{imp})}$ with extended-impurity-structure effects; percolation impurity behavior at $p = p_c^{(\mathrm{imp})}$; and, percolation spin behavior at $p = p_c$. Critical behavior of these types are expected to appear in dilute magnetic materials in the temperature range $|T - T_c(p)|/T_c(p) \leqslant (\Delta J/J_0)^{1/\varphi}$, determined by the value of the corresponding "crossover" exponent φ and ΔJ—the measure of randomness in the exchange interaction, for spin concentrations far from threshold values and in the region $|T - T_c(p)|T_c(p) \leqslant \{|p - p_c|/p_c\}^{1/\varphi}$ for $|p - p_c|/p_c \ll 1$. For Ising magnetic materials with $p_c^{(\mathrm{imp})} < p < 1\varphi = \alpha_{\mathrm{pure}} \simeq 0.11$, and for this reason the impurity behavior with the corresponding universal exponents should be observed in a narrow temperature range near $T_c(p)$ with "crossover" to exponents for homogeneous systems. For $p_c < p < p_c^{(\mathrm{imp})}$ "crossover" can be observed near the percolation threshold values. Far from these values either "crossover" is not observed or it can be manifested in the form of a transition between exponents for two types of impurity behavior. The results obtained in Ref. 7, where the dilute magnetic materials $Fe_pZn_{1-p}F_2$ with $p = 0.6$ and 0.5 were investigated precisely in the region $p_c < p < p_c^{(\mathrm{imp})}$ with $p_c = 0.25$, can be regarded as a unique experimental confirmation of this hypothesis. Critical exponents different from the exponents of a homogeneous system were obtained, but, to our surprise, no "crossover" to exponents of homogeneous critical behavior was observed.

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