

electron microscopy), and  $E_c = 30$  meV (estimated from a plot of  $\ln\sigma_L$  versus  $1/T$ ), we obtain  $2[(C_0/k)^{1/2}] \approx 120$  ( $^\circ\text{K}$ )<sup>1/2</sup>. The self-consistency of the model receives further confirmation in that the separation between the layers of grains calculated from the relation  $\mathcal{E}_0 = C_0/\Delta l e$  agrees with the mean value of  $s+d$  determined from the electron microscopy.

It is instructive to point out the similarities and the differences between Mott's model and ours. In Mott's model the density of charge carriers is assumed to be temperature independent, and the percolation paths for the charge carriers are determined by optimizing the mobility. In our model the charge carriers are thermally activated, tunneling occurs between nearest neighbors only, and the optimization is applied to the product of mobility and number density of charge carriers. The differences between charging energies in our model are analogous to the relative displacements of the energy levels for the localized states in Mott's model. However, in our case not only the differences but also the magnitudes of  $E_c$  play an important role. This is especially obvious in the high-field regime where the governing factor for field generation of charge carriers is the value of  $E_c$  rather than the differences in  $E_c$ .

To conclude, we would like to make the following comments: The field dependence,  $\exp(-\mathcal{E}_0/\mathcal{E})$ , of  $\sigma_H$  in the low-temperature regime is partially the result of high tunneling barriers in Ni-SiO<sub>2</sub>. In materials where the tunneling barrier is low, the high-field conductivity might well follow some other form of behavior (such

as the Frenkel-Poole effect). However, the relationship  $sE_c = \text{const}$  should still yield the  $\alpha = \frac{1}{2}$  behavior in the low-field regime. Attempts are being made to extend the concept of structural effects to other disordered materials. It is interesting to note that the inhomogeneous transport regime in disordered materials, recently treated by Cohen and Jortner,<sup>13</sup> bears close analogy to granular metals.

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## Scaling Theory for Metastable States and Their Lifetimes

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The response of the magnetization to a sudden reversal of the magnetic field is studied in the kinetic Ising model by means of computer experiments on square  $N \times N$  lattices. It is found that the nonequilibrium relaxation function fulfills a dynamic scaling hypothesis. The magnetization of the metastable state agrees with predictions of the cluster model and also with an analytic continuation of the linear-model equation of state.

Apart from systems where the mean-field approximation is valid,<sup>1</sup> no reliable predictions about the properties of metastable states exist. Monte Carlo calculations have been performed on the  $N \times N$  square kinetic Ising model,<sup>2,3</sup> and we

obtained the magnetization of the metastable states, their lifetimes, and the detailed nonequilibrium behavior.<sup>3</sup> Within the accuracy of the numerical calculations (roughly 1%) the results agree with simple scaling ideas.

Following Glauber<sup>4</sup> the dynamics of an Ising ferromagnet with nearest-neighbor interactions is described by a master equation for the probability distribution  $P(\mu_1, \dots, \mu_N, t)$  of the  $N$  spins  $\mu_j$  [ $\mu_j = \pm 1$ ], where a simple choice of a transition probability ensures development towards thermal equilibrium.<sup>4</sup> At time  $t=0$  the magnetic field  $H' > 0$  is changed by  $\Delta H$ , so that  $H' + \Delta H < 0$ . The resulting relaxation is described by the non-equilibrium relaxation function (NRF)<sup>1,5</sup>

$$\varphi_\mu^{\Delta H}(\vec{k}, t) = \frac{\sum_j \exp(i\vec{k} \cdot \vec{r}_j) [\langle \mu_j(t) \rangle - \langle \mu_j(\infty) \rangle]}{\sum_j \exp(i\vec{k} \cdot \vec{r}_j) [\langle \mu_j(0) \rangle - \langle \mu_j(\infty) \rangle]}, \quad (1)$$

$$\langle \mu_j(t) \rangle = \sum_{\{\mu_j\}} \mu_j P(\mu_1, \dots, \mu_N, t).$$

If  $\varphi_\mu^{\Delta H}(0, t)$  exhibits a long-living "flat" regime, we define a metastable state by a time average along this flat part.<sup>1</sup> This procedure does not prejudice the question of the existence of an essential singularity at the coexistence curve.<sup>6</sup>

The Monte Carlo simulations<sup>7-10</sup> on square  $N \times N$  lattices with periodic boundary conditions show that the NRF can be derived unambiguously (Fig. 1), and no dependence on  $N$  is found for the values  $|\Delta H|$  and  $\epsilon$  investigated<sup>11</sup> [see inset of Fig. 1]. For small values of  $|\Delta H|$  a flattening of the NRF does indeed occur,<sup>11</sup> a precursor of a metastable state develops, and for still smaller  $|\Delta H|$  the lifetime of the flat region is so large that it exceeds reasonable computing times (e.g.,  $10^4$  Monte Carlo configurations per spin). These flat regions with lifetimes considerably larger than the order-parameter relaxation time at equilibrium,  $\tau_{\delta\mu\delta\mu}$ ,<sup>9</sup> at that temperature, are

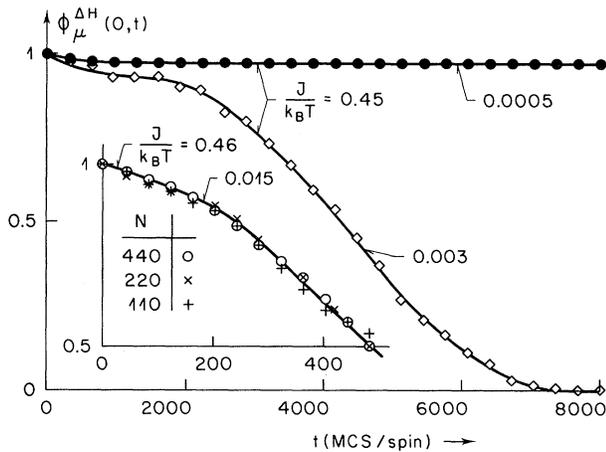


FIG. 1.  $\varphi_\mu^{\Delta H}(0, t)$  versus time at the temperature  $J/k_B T = 0.45$  ( $\epsilon \approx 0.0200$ ) for two values of the parameter  $-\mu_B H/k_B T$ . Inset,  $\varphi_\mu^{\Delta H}(0, t)$  for various  $N$  at  $-\mu_B H/k_B T = 0.015$ . In all cases  $H' = 0$  is used.

taken to characterize the metastable states.

Our hypothesis is that near the critical point [ $\epsilon = 1 - T/T_c \ll 1$ ,  $|H| = |H' + \Delta H| \ll k_B T/\mu_B$ ]

$$\varphi_\mu^{\Delta H}(\vec{k}, t) \equiv G(\epsilon, H, H', \vec{k}, t) = G_s(H_s, H_s', \vec{k}_s, t_s), \quad (2)$$

where  $H_s, H_s', \vec{k}_s$ , and  $t_s$  are scaled variables<sup>12,13</sup>:  $H_s = H\epsilon^{-\beta\delta}$ ,  $H_s' = H'\epsilon^{-\beta\delta}$ ,  $\vec{k}_s = \epsilon^{-\nu}\vec{k}$ ,  $t_s = t\epsilon^{\Delta\delta\mu\delta\mu}$ , with  $\beta$ ,  $\delta$ , and  $\nu$  standard exponents<sup>12</sup> and  $\Delta\delta\mu\delta\mu$  the exponent of the order-parameter relaxation time.<sup>9,14</sup> This implies that metastable states are consistent with static scaling and the coercive field  $H = H^*$  [where the flat part of the NRF starts becoming long lived, e.g.,  $\tau_R = 10^2 \tau_{\delta\mu\delta\mu}$ , where  $\tau_R \int_0^\infty \varphi_\mu^{\Delta H}(0, t) dt$  varies  $H^* \propto \epsilon^{\beta\delta}$ , and the magnetization at  $H^*$  varies  $\propto \epsilon^\beta$ ].

Let us evaluate the magnetization of the metastable states by two simple models, the linear-model equation of state<sup>15</sup> and the cluster model.<sup>6</sup> The linear-model equation of state leads to a cusp of the susceptibility  $\partial\chi_T/\partial H \propto -(H - H^*)^{-1/2}$  along a curve

$$\frac{\mu_B H^*}{k_B T_c} = -\frac{B}{C_+} \left[ \frac{\delta - 3}{\delta - 1} (1 - 2\beta) - 1 \right]^\beta \times \epsilon^{\beta\delta} \left( \frac{\delta - 1}{\delta - 3} \right)^{1/2} \left( \frac{1 - 2\beta}{2\beta} \right)^{\beta\delta} \frac{2}{\delta - 3}, \quad (3)$$

where  $B$  and  $C_+$  are critical amplitudes of magnetization and susceptibility, respectively, at equilibrium.<sup>12</sup> This description has been used in the discussion of experimental data on metastable He<sup>3</sup>.<sup>16</sup>

In contrast, Gaunt and Baker<sup>17</sup> found by series-extrapolation techniques a diverging susceptibility; their spinodal curve is<sup>17,18</sup>

$$\frac{\mu_B H^*}{k_B T_c} = -(0.39 \pm 0.2) \epsilon^{\beta\delta}. \quad (4)$$

If the general form of Schofield's parametric representation<sup>15</sup> is used, other types of singularities than the one given in Eq. (3) are possible.<sup>3</sup>

We also briefly discuss the approach based on Fisher's cluster model<sup>6</sup> where the average concentration of clusters with  $l$  reversed spins is taken to be

$$n_l \approx q_0 l^{-\tau} \exp \left[ -\frac{aJ\epsilon l^\sigma}{k_B T} - \frac{2H\mu_B l}{k_B T} \right], \quad l \rightarrow \infty, \quad (5)$$

where  $\sigma = \frac{8}{15}$ ,  $\tau = \frac{31}{15}$  (two-dimensional Ising model), and  $a$  and  $q_0$  are constants. Even for relatively small  $l$  this is a good representation of the actual cluster distribution.<sup>19</sup> The magneti-

zation is found from

$$\langle \mu \rangle = 1 - 2 \sum_{l=1}^{\infty} l n_l.$$

This relation cannot be used for  $H < 0$ , however, since then  $n_l$  has a minimum at the "critical cluster" size  $l^*$  and increases exponentially for large  $l$ . It has been suggested<sup>20,21</sup> that the supercritical clusters (with  $l > l^*$ ) are essentially characteristic of the new phase with reversed magnetization, and that one could estimate the magnetization in a metastable state from a relation where these clusters are removed, i.e.,

$$\langle \mu \rangle_{MS} = 1 - 2 \sum_{l=1}^{l^*} l n_l. \quad (6)$$

The resulting variation of  $\langle \mu \rangle_{MS}$  with  $H$  is smooth everywhere (except for small unphysical jumps at all integer values of  $l^*$  due to the cutoff). This approach does not yield the coercive field, unless one takes the extreme possible value  $l^* = l$ .<sup>6</sup>

In Fig. 2 we compare both predictions with the computer simulation. Equation (6) has been used down to  $l = 1$ , and  $a$  and  $q_0$  fitted to the critical amplitudes<sup>12</sup>; thus no adjustable parameters occur. In view of this fact the agreement between each prediction and the computer experiment is satisfactory. The critical fields  $H^*$ , where the lifetimes become large, nearly coincide with the linear-model prediction. The small deviations in the magnitude of  $\langle \mu \rangle$  are due to corrections to

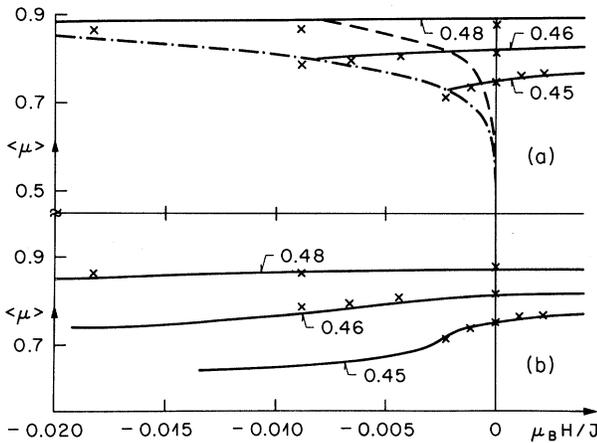


FIG. 2. Magnetization  $\langle \mu \rangle$  of metastable states versus the field. Full curves, (a) linear-model prediction and (b) cluster-model prediction; dash-dotted curve, linear-model prediction of the magnetization  $\langle \mu \rangle^*$  at the coercive field. Dashed curve,  $\langle \mu \rangle^*$  at the "pseudo-spinodal" predicted in Ref. 17. The parameter of the curves is  $J/k_B T$ . Crosses, the simulation results for  $N = 110$ .

scaling, as is evident for  $H = 0$  where the exact solution is available. The agreement with Eq. (6) is even better, since the droplet model accounts for a part of the correction terms. But Eq. (6) extends to unreasonably high values of  $-H$ ; clearly, the suggestion that  $l^* = 1$  determines  $H^*$  is not useful. Rather, one would like to identify  $H^*$  with the position of maximal slope of the  $\langle \mu \rangle$ -versus- $H$  curve.

The important fact is that both approaches yield results numerically rather consistent with each other and the computer simulation, and both approaches are easily generalized to other systems. Since both models are known to be a quite reasonable description of experimental data in equilibrium,<sup>15,22</sup> definite predictions about the metastable states of real systems seem feasible.

The cluster description<sup>6</sup> has the further merit that it can be generalized to provide a dynamic description of nucleation processes.<sup>23</sup> Similar results are derived from an approximate treatment of the master equation.<sup>3</sup> We summarize the essential points only.<sup>3</sup> In the linear approximation the time-dependent cluster distribution  $n_l'(t)$  obeys a continuity equation for the current  $J_l$ ,

$$\begin{aligned} \partial n_l'(t) / \partial t + \partial J_l / \partial l &= 0, \\ J_l &= (\bar{l})^2 R_l \partial n_l / \partial l + v_l n_l'(t), \end{aligned} \quad (7)$$

with some effective reaction rate  $R_l$  and effective size  $\bar{l}$  of the incorporated clusters in cluster reaction processes. Here  $v_l = -(\bar{l})^2 R_l \partial \ln(n_l) / \partial l$  is the growth velocity of a cluster. It is assumed<sup>23</sup> that the current in a metastable state is close to the steady-state solution of Eq. (7), which is approximated by<sup>23,3</sup>

$$J = \frac{\pi}{\sqrt{2}} (\bar{l})^2 n_l^* R_l \left( \frac{1}{n_l} \frac{\partial^2 n_l}{\partial l^2} \Big|_{l^*} \right)^{1/2}. \quad (8)$$

The magnetization is mainly decreased by the growth of already existing supercritical clusters, while new critical clusters are formed from the metastable phase at a nucleation rate  $J$ . Generalizing simple treatments of phase separation kinetics,<sup>24</sup> we write for the fraction  $X(t)$  of the new phase

$$dX = (1 - X) J V(t, t') dt', \quad (9)$$

where  $V(t, t')$  is the volume of a supercritical cluster at time  $t$  which originated at time  $t'$ . Calculating  $V$  from  $dV_l = v_l dt$ ,  $V_l \sim l$ ,  $R_l \sim l^{1-1/d}$  (in  $d$  dimensions) it is found that  $[l^* \sim (-a \sigma J \epsilon /$

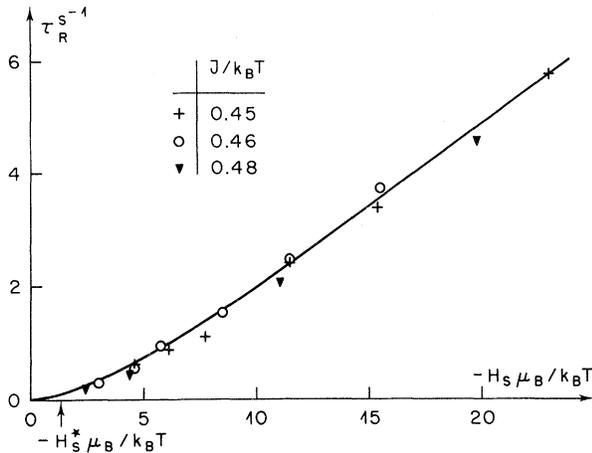


FIG. 3. Scaled inverse lifetime  $(\tau_R^s)^{-1} = \tau_R^{-1} \epsilon^{-\Delta_{\delta\mu\delta\mu}}$  versus scaled field. Full curve, nucleation-theory prediction, Eq. (10). The points are the computer-simulation results, derived from  $\tau_R \equiv \int_0^\infty \varphi^{\Delta H}(0, t) dt$ .  $H_s^*$  is the coercive field of the "linear-model" equation of state. Note that  $(\tau_{\delta\mu\delta\mu}^s)^{-1} \approx 6$ .

$$2H(\mu_B)^{1/(1-\sigma)} \sim H_s^{-1/(1-\sigma)} \epsilon^{-\gamma-\beta}$$

$$X(t) = 1 - \exp[-(t_s/\tau_R^s)^{d+1}],$$

$$(\tau_R^s)^{-1} = \text{const} \times (-2\mu_B H_s/k_B T)^{d/(d+1)} (l^* \epsilon^{\gamma+\beta})^\nu \\ \times \exp[2(1-\sigma)\mu_B H l^*/\sigma(d+1)k_B T], \quad (10)$$

$$y = \frac{\sigma/2 - 1/d - \tau}{d+1},$$

where the (unknown<sup>3</sup>)  $\epsilon$  dependence of  $(\bar{l})^2 R_f^*$  was adjusted to have agreement with Eq. (2). Figure 3 shows the excellent agreement obtained by this theory and the lifetimes found in the computer simulation. Only the constant factor in Eq. (10) is a fitted parameter. Thus dynamic scaling [Eq. (2)] is valid for nonequilibrium relaxation in the kinetic Ising model. Its validity near equilibrium was established previously.<sup>9,25-27</sup>

In conclusion, reproducible metastable states are found in the kinetic Ising model and interpreted in terms of the scaling hypothesis and the cluster picture. The results give no indication of the spinodal curve proposed by Gaunt and Baker for the Ising model,<sup>17,18</sup> but agree with the condensation description of Fisher<sup>6</sup> and Langer.<sup>21</sup> While the latter theories imply a smooth disappearance of the metastable states as  $-H$  is in-

creased, the linear-model representation suggests a possible existence of a coercive field, where  $\chi$  has a divergent slope, and  $\varphi_\mu^{\Delta H}(\vec{k}, t)$  can have some singularity (e.g.,  $\tau_R^s$  has a divergent slope there). Unfortunately, the precise analytic behavior<sup>3</sup> at the coercive field  $H^*$  cannot be investigated by the present techniques.

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## Ruderman-Kittel-Kasuya-Yosida Spin Polarization in a Strongly Perturbed Medium and Applications to Hyperfine Fields

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Hyperfine fields at normal impurities in ferromagnetic metals and alloys vary systematically in sign and magnitude. These results are explained by an extension of Ruderman-Kittel-Kasuya-Yosida spin polarization to the case of an electron gas with a strong local perturbation.

The hyperfine field at *sp* impurities in the ferromagnetic metals has attracted considerable attention since it was shown experimentally that the fields were systematically negative for elements in the first half of an *sp* series and positive for elements in the second half. It has been a bone of contention whether the observed positive fields can be explained by purely conduction-electron effects<sup>1,2</sup> or whether it is necessary to invoke a direct overlap mechanism between host *d* orbitals and *s* states on the impurity.<sup>3,4</sup>

Recently, a number of results have been obtained<sup>5-11</sup> for the hyperfine fields at the *sp* sites *Y* in the Heusler alloys  $X_2MnY$ , Table I. Although the results are rather incomplete, it appears that a crossover occurs from negative to positive fields when the element *Y* is in the middle of the *sp* series, as for *sp* impurities in Fe or Ni hosts.<sup>12</sup> For the Heusler compounds the *Y* site has no magnetic nearest neighbors, so any direct overlap would be small; the change in sign of the field appears to be a purely conduction-electron effect.

The results can be compared with the model of Caroli and Blandin<sup>13</sup> which has been widely used in interpreting experimental data. In this model, the conduction-electron band is taken as free-electron-like with an effective number of electrons per atom equal to the average over all sites. A *d* resonance at each Mn site leads to

spin-density oscillations, and summing over the contributions from different Mn sites around a given nonmagnetic atom leads to a prediction for the hyperfine field at that site. While predictions for *X* sites are good, fields at *Y* sites are generally expected to be negative and to vary little with the effective charge on the *Y*-site element, in disagreement with experimental results, Table I.

A weakness of the model is that the charge screening at the nonmagnetic site is not included explicitly<sup>14</sup>; this has little importance for nuclei such as Cu on *X* sites, but may be very important for *Y* sites where the effective charge is large. Here we calculate the two-impurity problem in order to show the effect of the charge screening at the nonmagnetic site on the polarization at that site. This calculation is an extension of the Ruderman-Kittel-Kasuya-Yosida (RKKY) approach to an electron gas with a strong perturbation, in the same spirit as the calculation of Daniel and Friedel.<sup>1</sup>

The conduction electrons will be considered as free-electron-like with Fermi wave vector  $k_F$ . A normal impurity at the origin has a spin-independent spherical potential  $V_0(r)$  [with  $V_0(r) = 0$  for  $r > r_0$ ] giving rise to phase shifts  $\delta_l$ . This potential will not be treated within the Born approximation, but its effect will be treated exactly