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# Experimental verification of activated critical dynamics in the $d=3$ random-field Ising model

A. E. Nash, A. R. King, and V. Jaccarino
*Department of Physics, University of California, Santa Barbara, California 93106*
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ac susceptibility studies of the $d=3$ random-field Ising-model (RFIM) system $Fe_{0.46}Zn_{0.54}F_2$ in the $5\times10^{-3}\leq\omega/2\pi\leq10^5$ Hz frequency range show the $\omega$ scaling of the peak value $[\chi_c'(\omega)]_p$ and the characteristic rounding temperature $t^*(\omega)$ are consistent with activated, but not conventional, dynamic scaling. We find $[\chi_c'(\omega)]_p\sim\log|\log(\omega/\omega_0)|$ and $t^*(\omega)\sim|\log(\omega/\omega_0)|^{-1/y}$ with $y=1.05\pm0.16$ and $\omega_0\simeq10^8$ $s^{-1}$ in agreement with the random-exchange Ising-model (REIM) dynamic frequency at the REIM-to-RFIM crossover. The relation between these results and the Villain-Fisher theories is discussed.

As the understanding of the $d=3$ random-field Ising model (RFIM) has evolved over the past decade, it has become clear that the dynamical behavior in the vicinity of the phase transition $T_c(H)$ plays a crucial role in determining the RFIM's rather unusual critical properties. Specifically, the critical slowing down is so extreme that the characteristic time scale of these systems exceeds even the longest of laboratory time scales as $T\rightarrow T_c(H)$. This attribute of the RFIM was predicted by heuristic theories of Villain[1] and Fisher[2] (VF) and experimentally evidenced and quantified in ac susceptibility[3] and neutron-scattering studies[4] of the prototypical RFIM system $Fe_xZn_{1-x}F_2$; a randomly diluted uniaxial antiferromagnet in a uniform applied field.

The VF theories incorporate an *activated* dynamics in which the characteristic time $\tau$ for a fluctuation on a scale of the correlation length $\xi$ grows as $\tau\sim\exp(\xi^\theta)$, whereas in a "conventional" dynamic approach $\tau\sim\xi^z$. The previous studies[3] on $Fe_{0.46}Zn_{0.54}F_2$ were not able to distinguish activated from conventional behavior that had an anomalously large value of $z$. The present work has as its goal the resolution of this issue through the extension of the frequency range over which the dynamics are probed. While our results show that an activated and *not* a conventional dynamics approach is required, the experimentally determined value of $\theta$ is not consistent with *all* of the VF scaling relations, when subject to the Schwartz-Soffer inequality[5] and our existing knowledge of the exponents.

The $Fe_{0.46}Zn_{0.54}F_2$ crystal used is the same one employed in previous susceptibility $[\chi(\omega)]$ measurements[3] and birefringence $(\Delta n)$,[6] capacitance,[7] neutron-scattering,[8] and neutron spin-echo[9] experiments. Measurements of $\chi'(\omega)$ as a function of temperature $T$ by a mutual-inductance technique at frequencies 1 kHz $\leq\omega/2\pi\leq$ 100 kHz were made with an ac field of $h_{ac}\lesssim1$ Oe. $\chi'(\omega)$ and $\chi''(\omega)$ were also studied as a function of $T$ by optical Faraday rotation (FR) at $5\times10^{-3}$ Hz $\leq\omega/2\pi\leq2\times10^{-2}$ Hz with $h_{ac}=50$ Oe. A resolution of $\delta\Theta=0.002°$ was obtained for the FR angle $\Theta$ using a sensitive compensation circuit[10] involving a photoelastic modulator, lock-in detection, and a feedback-controlled, rotating analyzer. After dc magnetization compensation at each $T$, the lock-in output was Fourier transformed to provide $\chi'(\omega)$ and $\chi''(\omega)$. All measurements were made in an applied field of $H=10$ kOe. The temperature was stabilized to $\pm1$ mK by means of a field-insensitive carbon-glass thermometer and an ac resistance bridge. Uniform "dc" magnetization measurements were also made (similar to those made in $Fe_{0.47}Zn_{0.53}F_2$)[11] and will be reported elsewhere.

A typical $\chi'(\omega)$-vs-$T$ result at $\omega/2\pi=0.02$ is shown in Fig. 1. No observable shift in the peak temperature was seen as a function of $\omega$. As before, any dynamical critical-behavior analysis requires the separation of the smooth $\omega$-independent background [effectively $\chi'(\omega=\infty)$] from the critical part, $\chi_c'(\omega)$. We know the static divergence, $\chi'(\omega=0)$, must be the same as the magnetic specific heat $C_m=(A^{\pm}/\alpha)(|t|^{-\alpha}-1)$, where $A^+$ $(A^-)$ is the amplitude above (below) the transition, $t$ is the reduced temperature relative to $T_c(H)$, and $\alpha$ is the specific-heat exponent. $C_m$ has been found[12] to have a symmetric, logarithmic divergence at $T_c(H)$, where $A^+/A^-\simeq1$ and $\alpha=0.00\pm0.03$. The appropriate $\chi_c'(\omega)$ was constructed as reported previously.[3] Exactly the *same* background function[3] was subtracted from the data for all $\omega$. The resulting background-corrected data is shown in the $\chi_c'(\omega)$ versus $t$ plot of Fig. 2 for three values of $\omega/2\pi$.

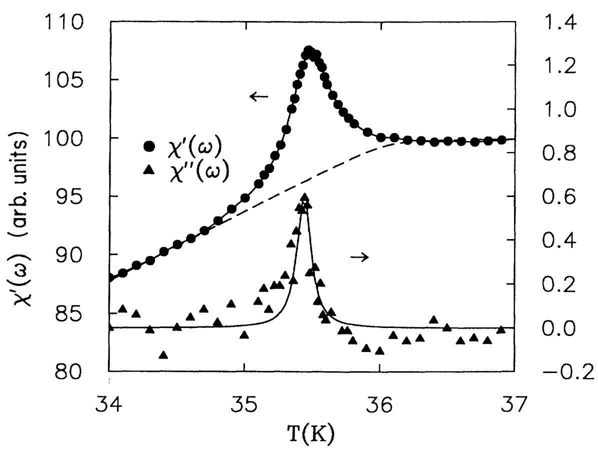


FIG. 1. The real and imaginary parts of $\chi(\omega)$: $\chi'(\omega)$ and $\chi''(\omega)$, for $\omega/2\pi=0.02$ Hz vs $T$. The dashed line is the background contribution as described in the text. Note $\chi''\ll\chi'$ and that $\chi''$ is vanishingly small outside the reduced temperature region $|t|<5\times10^{-3}$. The solid lines are guides to the eye.

 

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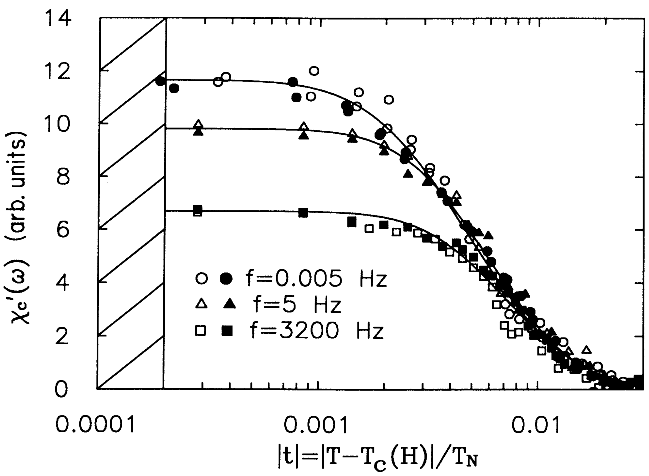


FIG. 2. $\chi'(\omega)$ vs $\log_{10}|t|$, the reduced temperature, for just three frequencies; $\omega/2\pi=0.005$, 5, and $3.2\times10^3$ Hz, after background subtraction, as described in the text. The open and solid symbols refer to $T<T_c(H)$ and $T>T_c(H)$, respectively. Rounding of the transition due to the concentration gradient occurs only within the shaded area; i.e., $|t|<2\times10^{-4}$.

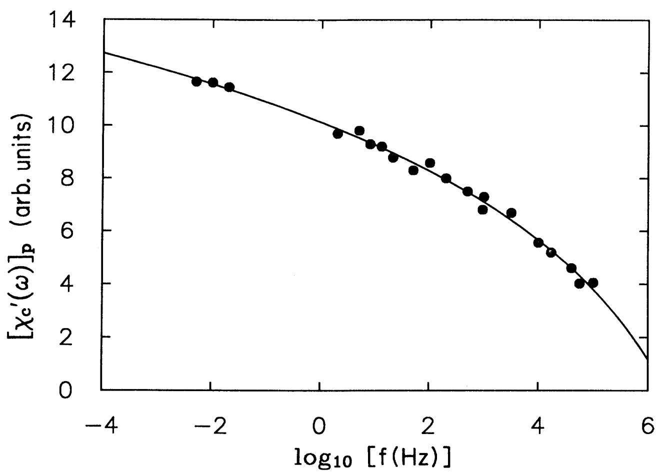


FIG. 3. Peak value of background corrected susceptibility, $[\chi_c'(\omega)]_p$ vs $\log_{10}\omega/2\pi$. The solid line is a best fit to activated scaling $[\chi_c'(\omega)]_p \sim \ln|\ln(\omega/\omega_0)|$ with $\omega_0=5.9\times10^8$ s$^{-1}$. Errors are on the order of the size of the data points. Conventional scaling would have lead to a linear relation between $[\chi_c'(\omega)]_p$ and $\log_{10}\omega/2\pi$.

The two important features of $\chi_c'(\omega)$ are the $\omega$ scaling of the peak height $[\chi_c'(\omega)]_p$ and the rounding temperature $t^*(\omega)$ at which the system departs from equilibrium on a time scale $\tau=\omega^{-1}$. Since the amplitude ratio $A^+/A^-$ should be unity in the RFIM critical region, it follows that the experimentally determined $[\chi_c'(\omega)]_p \equiv \chi_c'(\omega)_{\max}$ which is plotted versus $\log_{10}\omega$ in Fig. 3. The previous method[3] of determining $t^*(\omega)$ depends on the ability to determine the amplitude $A^\pm$ of the $\log|t|$ divergence. As seen in Fig. 2, the amplitude $A^\pm$ is difficult to determine from the data, particularly at the higher $\omega$, because of the extensive region of $t$ in which dynamical effects dominate. Instead, we make a scaling analysis which makes no *a priori* assumptions about $A^\pm$.

In the absence of a detailed theory, we adopt a scaling analysis similar to that used for the $d=2$ RFIM specific heat[13] (where the leading divergence was also $\ln|t|$). Generalizing the scaling prediction to the case where $\alpha \to 0$ we take $\chi_c'(\omega)$ to have the form

$$\chi_c'(\omega)=g[t/\Omega(\omega/\omega_0)]-A^*\ln|\Omega(\omega/\omega_0)|+B\,, \qquad (1)$$

where $g(x)$ is a scaling function, $\Omega$ is a function only of the ratio of $\omega$ to some characteristic frequency $\omega_0$ of the system, and $A^*$ and $B$ are constants. As can be seen from Eq. (1), $t^*(\omega) \propto \Omega(\omega/\omega_0)$ and

$$[\chi_c'(\omega)]_p=-A^*\ln|\Omega(\omega/\omega_0)|+B^*$$

where $B^*=g_{\max}+B$.

Activated dynamical behavior is a property of systems that have energy barriers between states of similar energy. In this case, the time for a fluctuation on a scale of $\xi$ should grow exponentially with $\xi$. We generalize this to have the divergent form $\tau=\tau_0\exp^{E/t^y}$ where $\tau_0^{-1}$ is some characteristic exchange frequency, $E$ is a constant, and $y$ is a parameter to be determined. (The simple Vogel-Fulcher[14,15] law has $y\equiv1$, whereas in the VF (Refs. 1 and 2) theories $y=\theta\nu$, with $1\le\theta\le d-1$ and $\nu$ is the RFIM correlation length exponent.) It follows that $\Omega(\omega/\omega_0) \propto |\log(\omega/\omega_0)|^{-1/y}$ in which case activated dynamics would predict, in the limit $\alpha\to0$,

$$[\chi_c'(\omega)]_p=(A^*/y)\ln|\ln(\omega/\omega_0)|+B^*\,. \qquad (2)$$

A fit of Eq. (2) to the peak height data, with reduced $\chi_\nu^2=1.02$, yields a value of $2.0\times10^8$ s$^{-1}\le\omega_0\le2.0\times10^9$ s$^{-1}$ and is shown in Fig. 3. We note

$$\chi_c'(\omega)-[\chi_c'(\omega)]_p=g[t/t^*(\omega)]-g_{\max}\,, \qquad (3)$$

so that $\chi_c'(\omega)-[\chi_c'(\omega)]_p$ for each frequency measured will collapse onto the lowest frequency $\omega_L$, at which measurements were made, in the RFIM critical region, if $t$ is rescaled by the normalized ratio $r(\omega)=t^*(\omega)/t^*(\omega_L)$. This procedure is illustrated in Fig. 4. The value of $r(\omega)$ for each $\omega$ is determined by the best collapse of the data at $\omega$ onto that at $\omega_L$. For activated scaling, $t^*(\omega) \propto |\ln(\omega/\omega_0)|^{-1/y}$ so that

$$r(\omega)=t^*(\omega)/t^*(\omega_L)=\left|\frac{\ln(\omega/\omega_0)}{\ln(\omega_L/\omega_0)}\right|^{-1/y}. \qquad (4)$$

A fit of the $r(\omega)$ data to Eq. (4) with reduced $\chi_\nu^2=3.09$, gives $y=1.05\pm0.16$ and $10^7$ s$^{-1}\le\omega_0\le2.5\times10^8$ s$^{-1}$ consistent with the value of $\omega_0$ obtained from our fit of $[\chi_c'(\omega)]_p$. The data and best fit is shown in Fig. 5.

When $\omega=0$, one expects $\chi_c'(\omega)\propto\ln|t|$; hence $g(x)\sim -A\ln|x|$ for $|x|\gg1$. It follows from Eq. (1) that as $\omega\to0$, $\chi_c'(t,\omega\to0)=-A\ln|t|+B$ where $A^*=A$. In the previous analysis[3] $t^*(\omega)$ was defined $[\chi_c'(\omega)]_p=-A\times\ln|t^*(\omega)|+B$, where the estimate of $A$ limits the ability to determine $y$. Our value of $A^*$ from the fit of the data to Eq. (2) implies the previous[3] estimates of the amplitude of the best symmetric $\ln|t|$ divergence, and subsequently $y$, may have been in error by only 20%.

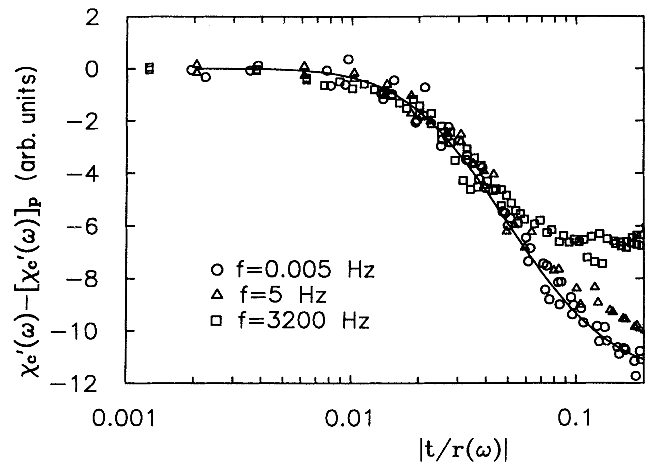


FIG. 4. $\chi_c'(\omega)-[\chi_c'(\omega)]_p$ vs $|t/r(\omega)|$ at the same three frequencies of Fig. 2, showing the collapse of the scaled data in the RFIM region. The solid line is a guide to the eye. Departures from scaling occur for scaled reduced temperature above the REIM-to-RFIM crossover.

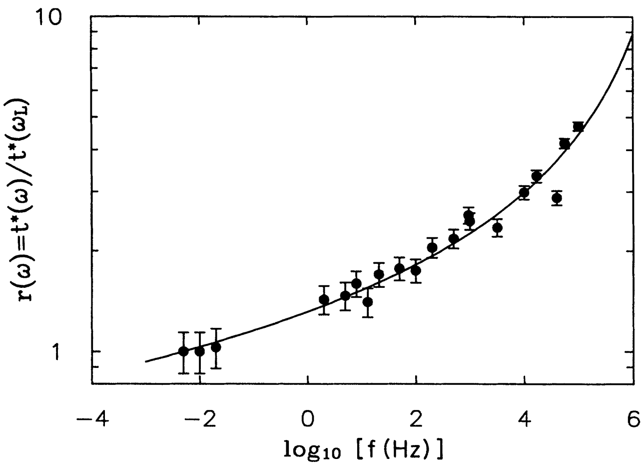


FIG. 5. Normalized dynamic rounding temperature ratio $r(\omega)=[t^*(\omega)/t^*(\omega_L/2\pi=0.005\text{ Hz})]$, as described in the text, vs $\log_{10}\omega/2\pi$. A fit to activated scaling $r(\omega)=|\ln(\omega/\omega_0)/\ln(\omega_L/\omega_0)|^{-1/y}$ with $y=1.05$ and $\omega_0=5.5\times10^7\ \text{s}^{-1}$ is also shown as the solid line. Conventional scaling would have lead to a linear relation between $r(\omega)$ and $\log_{10}\omega/2\pi$.

One might wonder why the values of $\omega_0$ obtained from the fits of $[\chi_c'(\omega)]_p$ and $r(\omega)$ are so much less than the exchange frequency of pure $FeF_2$; $\omega_0\approx10^{12}\ \text{s}^{-1}$. The time scale for fluctuations is set by the reduced temperature $t_{cr}$(H) at the crossover boundary between the random-exchange Ising model and the random-field Ising model using the relation

$$\tau_{cr}(H)=\tau_0 t_{cr}^{-\nu z}(H)\,, \tag{5}$$

where $\tau_{cr}(H)$ and $\tau_0$ are the characteristic time scales in the RFIM and REIM regions, respectively, where here $\nu$ and $z$ refer to the REIM correlation length and dynamical critical exponents, respectively. From neutron spin-echo[9] and high-precision birefringence measurements[6] we estimate $7\times10^7\ \text{s}^{-1}\leq\omega_0\leq2.9\times10^8\ \text{s}^{-1}$, which agree with the values obtained from the fits of $[\chi'(\omega)]_p$ and $t^*(\omega)$. Therefore, the $\omega_0$ obtained from our fit *does* match the expected value at the crossover boundary. Note that different choices of background functions, while affecting the $|t|>0.01$ region, in no sense affects the $\omega$ dependence of $[\chi_c'(\omega)]_p$ or $t^*(\omega)$.

In conventional dynamics, $\tau$ grows with $\xi$ as $\tau\sim\xi^z$; hence $\Omega(\omega/\omega_0)\propto\omega^{1/z\nu}$ and, in the limit $\alpha\to0$, it would predict $[\chi_c'(\omega)]_p\sim|\log(\omega)|$ and $t^*(\omega)\propto\omega^{1/z\nu}$. If this were so, the plots of the measured $[\chi_c'(\omega)]_p$ and $t^*(\omega)$ vs $\log_{10}\omega/2\pi$, in Figs. 3 and 5, respectively, would lie along straight lines. This is clearly not the case. There is another crucial failure of the conventional dynamics description. As Fig. 1 shows, the imaginary part $\chi''(\omega)$ is roughly 10 times *smaller* than the critically divergent real part $\chi_c'(\omega)$. This is indicative of a wide distribution of relaxation times, whereas the conventional dynamical approach is characterized by a single relaxation time (exponential decay of the correlation function).

How does the experimentally determined value of $y=1.05\pm0.16$ compare with the theoretical predictions for an activated dynamics? First, it is interesting to note that it is consistent with the simple Vogel-Fulcher[14,15] law ($y\equiv1$). In the VF model[1,2] of activated dynamics $y\equiv\theta\nu$. The parameter $\theta$ appears in a violated hyperscaling relation

$$(d-\theta)\nu=2-\alpha \tag{6}$$

and is unique in that it relates static and dynamic exponents.[16] Using the experimental value of $y$ and $\nu=1.0\pm0.15$ (Ref. 17) (or $1.0\pm0.1$) (Ref. 18) one obtains $\theta=1.05\pm0.22$ (or $1.05\pm0.18$). This value of $\theta$, when combined with the above value(s) of $\nu$, and the experimental value of $\alpha=0.00\pm0.03$,[12] satisfies Eq. (6).

However, there is an additional VF scaling relation; $\theta=\eta-\tilde{\eta}$ with $\eta$ and $\tilde{\eta}$ the correlation function and disconnected susceptibility exponents, respectively. Using the Monte Carlo result $\tilde{\eta}=-0.9$ (Ref. 18) and the inequality $\eta>(4-d)/2^5$ one finds $\theta\gtrsim1.5$ which does *not* agree with the value 1.05 given above. Furthermore, the value of $\theta\gtrsim1.5$ does *not* satisfy Eq. (6). It is clear the experimental results on $\alpha$ and $\theta$ do *not* agree with the equilibrium theory predictions[1,2,16] in which the static and dynamic critical behavior is controlled by a zero temperature fixed point. But it is interesting to note that all of the measured experiments *are* in agreement with Shapir's phenomenological theory[19] in the "local response" regime.

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