PHYSICAL REVIEW B VOLUME 44, NUMBER 9 1 SEPTEMBER 1991-I

Ultrasonic investigation of critical behavior in the random-field Ising system $Dy(As_xV_{1-x})O_4$

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(Received 21 March 1991)

Elastic-constant measurements, made with the use of a high-precision ultrasonic interferometer, have allowed determination of the susceptibility critical exponent γ in the random-field Ising system $Dy(As_xV_{1-x})O_4$. For pure $DyVO_4$ a value $\gamma = 1.15 \pm 0.05$ was obtained, which is between the values $\gamma = 1$ for a mean-field system and $\gamma = 1.25$ for a three-dimensional Ising system with nearest-neighbor interactions, and consistent with conclusions reached by previous authors that there is a strong short-range component to the exchange interactions. Random-field samples yielded a significantly higher value, specifically $\gamma = 1.79 \pm 0.07$, consistent with the two-dimensional nearest-neighbor Ising value $\gamma = \frac{7}{4}$ and with results of simulations by other authors, but in disagreement with certain scaling-theory predictions. This is the most reliable value obtained thus far for γ in a random-field Ising system in three dimensions. Results of optical-birefringence measurements on the ultrasonics samples, along with numerical simulations, show that the measurement of γ was not significantly affected by arsenic-concentration inhomogeneities.

I. INTRODUCTION

Since the pioneering work of Imry and Ma,[1] Ising systems in which a site-random field (RF) couples to the order parameter have been the subject of considerable theoretical and experimental study.[2] RF suppression of long-range order leads to lower transition temperatures, new critical behavior, and the formation of metastable domains in the low-temperature phase. This work is an experimental investigation of the critical behavior, in particular of the isothermal susceptibility exponent γ, for the random-field Ising model (RFIM).

There is no exact physical realization for RF's in Ising ferromagnets, and most experimental studies have been of dilute antiferromagnets (DAF's) in a uniform applied field, in which case a RF which couples to the staggered magnetization is generated, proportional to the applied field.[3] A previous investigation of static RFIM critical behavior in DAF's has found $\alpha = 0.00 \pm 0.03$ for the specific-heat critical exponent, determined from birefringence experiments[4] on $Fe_{0.46}Zn_{0.54}F_2$. In addition, the values $\gamma = 1.75 \pm 0.2$, $\nu = 1.0 \pm 0.15$ have been measured[5] for the static susceptibility and correlation-length exponents in $Fe_{0.6}Zn_{0.4}F_2$, although these values should be viewed with caution since the (staggered) susceptibility and correlation length in an antiferromagnet can be obtained only from neutron scattering measurements of the growth of a superlattice peak and are sensitive to the functional form used to describe the line shape, for which there is no rigorous theory currently available.[6]

The particular system under study in this work is the random-strain compound $Dy(As_xV_{1-x})O_4$—a mixture of the isomorphic compounds $DyVO_4$ and $DyAsO_4$, each of which undergoes a cooperative Jahn-Teller transition from tetragonal to orthorhombic symmetry, at temperatures $T_D = 14.6$ K and 11.4 K, respectively.[7] In this system the interactions between the Dy^{3+} ions can be described by an effective Ising pseudospin Hamiltonian in which the order parameter is proportional to a B_{1g} lattice strain.[8] The mismatch of $DyVO_4$ and $DyAsO_4$ lattice parameters[9,10] generates a random strain field, one component of which has the correct symmetry to couple to the order parameter.[11,12] The susceptibility χ is the inverse of an elastic constant — specifically $c_S \equiv \frac{1}{2}(c_{11} - c_{12}) \propto \chi^{-1}$, which vanishes at the second-order phase transition[13]—and can therefore be measured directly in an ultrasonic velocity experiment, from which γ can be determined from $c_S \propto \epsilon^\gamma$, where $\epsilon = (T - T_D)/T_D$ is the reduced temperature. Although the critical behavior of pure $DyVO_4$ is believed to be ultimately classical because of long-range coupling to a uniform strain, this behavior is not observed for experimentally accessible temperatures close to T_D, owing to strong short-range interactions.[7,14] These short-range interactions are also expected to dominate the long-range strain coupling in the mixed compounds $Dy(As_xV_{1-x})O_4$, allowing meaningful (although perhaps not exact) comparison of the observed critical exponents in $Dy(As_xV_{1-x})O_4$ with the predictions of the usual nearest-neighbor RFIM. We note that although there is evidence that dilution of the magnetic species may impede the observation of equilibrium critical behavior in DAF's,[15] there is no such dilution in $Dy(As_xV_{1-x})O_4$ since the interactions driving the transition are between the Dy^{3+} ions, so only the

RF's, and not the exchange interactions, should depend on x.

Metastability and the behavior of domains in this system have been discussed elsewhere,[12,16,17] as have preliminary ultrasonics results.[17,18]

II. ULTRASONIC VELOCITY MEASUREMENTS

The elastic constant $c_S = \rho(v_S)^2$, where ρ is the density, can be determined directly from the velocity v_S of shear waves propagating in the [110] direction;[19,20] however, large ultrasonic attenuation near T_D prohibits such measurements in the critical region.[19] We avoided this problem by measuring the velocity v_L of longitudinal waves propagating in the [100] direction, which yields

$$\rho(v_L)^2 = c_{11} = \tfrac{1}{2}(c_{11} + c_{12}) + \tfrac{1}{2}(c_{11} - c_{12}),$$

from which the temperature-independent $c_{11}^0 = \frac{1}{2}(c_{11} + c_{12})$ can be subtracted,[21] leaving $c_{11} - c_{11}^0 = c_S \propto \epsilon^\gamma$.

Changes in sound velocity as a function of temperature were measured very precisely with an ultrasonic interferometer, using a pulse-echo technique.[22] Radio-frequency pulses were generated electronically, transduced to ultrasonic signals, and then passed through a quartz delay rod into the sample, which had flat and parallel [100] faces, polished to a fraction of an ultrasonic wavelength. The echoes which resulted from multiple reflections from these faces were retransduced to electrical signals and mixed with a variable-phase reference signal that was coherent with the initially generated pulse. The reference phase was then adjusted to give a destructive interference condition at the center of the first "sample" echo. A change in sample sound velocity caused a change in echo arrival time and could therefore be calculated from the change in phase required to maintain destructive interference. In practice, this method allowed detection of phase shifts as small as $\sim 5 \times 10^{-4}$ cycle, for reasonably noise-free 30-MHz pulses and excellent frequency stability, giving a sensitivity to changes in velocity of about one part in 10^5 for typical samples used in these experiments.

These ultrasonic velocity experiments required large homogeneous single crystals, the growth and As concentration measurement of which have been described elsewhere.[9] Samples used for [100] measurements had $x = 0$, 0.154 ± 0.010, 0.164 ± 0.010, and 0.984 ± 0.001. We also used a [110] sample with $x = 0.154 \pm 0.010$. The thickness d along the propagation direction needed to be sufficient to separate the echoes obtained from multiple reflections in the sample. The round-trip propagation time was typically $t_0 \sim 1$ μs, and a pulse duration of 0.4 μs provided adequate separation while maintaining reasonable spectral purity at the peak (necessary for the interferometry). The sample was bonded to the quartz rod by a thin (30–50 μm) indium foil with very lightly greased surfaces. The grease allows good ultrasonic transmission, and the indium provides strain relief from differential thermal contraction during cooling.

The absolute propagation time t_0 was measured at room temperature by feeding the reflected signal into both channels of a variable-delay oscilloscope and delaying one channel for overlap of two of the multiple echoes.[23] In order to correct for possible phase shifts associated with the pulse reflections, the delay was measured at several frequencies from 20 to 300 MHz. A linear relationship was observed between the time delay and f^{-1}, implying a shift in phase that was independent of f; we corrected for this by extrapolating to $f^{-1} = 0$. The value for t_0 thus obtained was then used to determine the absolute velocity, $v_0 = 2d/t_0$.

Once t_0 was determined, the interferometer was used to monitor the phase of the first sample echo as a function of temperature down to 4.2 K, with particular attention paid to measurements near the phase transition. Velocities were then calculated from $v = 2d/(t_0 + \Delta t)$, where Δt is the measured reference phase shift (in time units) relative to t_0. Results for $c_{11} = \rho v^2$ for the four [100] samples are shown as a function of temperature in Fig. 1, which clearly illustrates the softening of c_S associated with the transition. For all samples studied c_{11} decreases continuously to a minimum value at $T = T_D$, implying that the phase transition is second order in the RF samples as well as in pure DyVO$_4$. As the strength of the RF is increased by increasing the concentration of the minority species (As or V), T_D becomes more severely suppressed and the transition becomes broader. Some of the results of this work are summarized in Table I.

TABLE I. Summary of results, for different arsenic concentrations x, showing: sample density ρ (from Ref. 9), sample thickness d, room-temperature propagation time t_0 for a round trip though the sample, room-temperature sound velocity v(RT)$= 2d/t_0$ and elastic constant c(RT)$= \rho[v(\mathrm{RT})]^2$, transition temperature T_D, sound velocity v_{11}^0 and elastic constant c_{11}^0 at $T = T_D$ (for [100] samples), and susceptibility critical exponent γ.

x	ρ (g/cm^3)	d (mm)	t_0 (ns)	v(RT) (km/s)[a]	c(RT) (GPa)[b]	T_D (K)	v_{11}^0 (km/s)	c_{11}^0 (GPa)	γ
0	5.7168 (8)	2.900 (2)	896.27 (19)	6.471 (6)	239.4 (5)	14.075 (10)	5.211 (5)	155.2 (3)	1.15 (5)
0.154 (10)	5.811 (6)								
[100]		2.926 (2)	909.54 (10)	6.434 (5)	240.5 (6)	7.95 (2)	5.177 (5)	155.8 (5)	1.80 (6)
[110]		1.500 (2)	739.79 (9)	4.055 (6)	95.6 (4)				
0.164 (10)	5.820 (6)	2.915 (2)	900.33 (27)	6.475 (6)	244.0 (5)	5.60 (3)	5.206 (6)	157.8 (5)	1.79 (7)
0.984 (5)	6.332 (4)	1.939 (2)	610.8 (5)	6.349 (12)	255.2 (11)	9.07 (3)	5.128 (15)	166.5 (11)	1.76 (15)

[a] $v = v_L$ for the [100] samples, and $v = v_S$ for the [110] sample.

[b] $c = c_{11}$ for the [100] samples, and $c = \frac{1}{2}(c_{11} - c_{12})$ for the [110] sample.

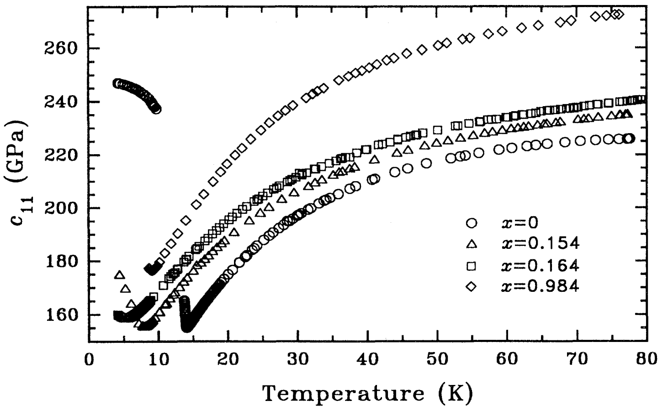

FIG. 1. Elastic constant c_{11} vs temperature, for all four [100] samples. The data for the $x = 0.984$ sample have been displaced vertically upward by 10 GPa for clarity.

Since the main aim of these measurements is to determine static critical exponents, we must ensure that the transition is not smeared by noncritical effects, especially if such smearing would prevent the measured values of the soft-mode elastic constant c_s from vanishing at $T = T_D$. Since T_D is strongly dependent on arsenic concentration x, one source of rounding of the transition could be inhomogeneity in x in a RF sample. This point has been emphasized by King, Belanger, Ferreira, and Jaccarino[24,25] (hereafter referred to as KBFJ) with reference to DAF's. We have used a technique similar to that of KBFJ to determine the variation in x in our samples from c-a birefringence measurements. These measurements, details of which are given in a previous paper,[9] exploit the sensitivity of the birefringence Δn_{ca} to x by converting sensitive measurements of the spatial variation in Δn_{ca} to variations in x and hence in T_D. Variations in concentration as small as 10^{-5} could be resolved using this technique, although uncertainty in sample parallelism prevented this ultimate sensitivity from being achieved in practice. Results for the RF samples are summarized in Table II, which gives for each sample the total variation in apparent birefringence[26] $\delta(\Delta n_{ca})$, As concentration δx, transition temperature δT_D, and reduced temperature $\delta\epsilon$. The range of transition temperatures influencing the ultrasonic velocity was further reduced in each sample by cutting the indium bonding layer to transmit sound only through the region of greatest uniformity, which corresponds to the optically clearest region. It is also possible that much of the observed "inhomogeneity" in our samples is due to thickness variation, although small oscillations in the apparent birefringence[9] in our most homogeneous sample give a lower bound for $\delta\epsilon$ of $\sim 7 \times 10^{-4}$. Thus the values for δT_D relevant to our ultrasonic velocity measurements are probably rather less than those given in Table II, which at any rate are too small to account for the fact that the minimum in c_{11} is less sharp in the RF samples than in pure $DyVO_4$ (Fig. 1), especially since the broadest transition occurs in the most homogeneous RF sample.

Another potential source of rounding of the minimum in c_{11} could be dynamic nonequilibrium effects due to extreme slowing down of critical fluctuations as the transition is approached.[27] Such dynamic rounding has been observed in ac susceptibility experiments[28] on the DAF $Fe_{0.46}Zn_{0.54}F_2$, which indicate a rounding onset temperature and a suppression of the full static response that is weakly dependent on frequency. Investigation of dynamic rounding in $Dy(As_xV_{1-x})O_4$ requires comparison of measurements made at a range of ultrasonic frequencies. Preliminary results of such measurements[29] at frequencies from 25 to 120 MHz suggest that for the strongest RF sample, $x = 0.164$, the minimum in c_{11} becomes shallower as f is increased above 30 MHz, indicating that dynamic critical effects could be affecting the present data for this sample. However, no such frequency dependence is observed in the somewhat weaker RF sample, $x = 0.154$, between 10 and 70 MHz, so that in our experimentally accessible range of temperatures near T_D the static critical behavior of this sample is not obscured by the dynamics.

III. SUSCEPTIBILITY CRITICAL EXPONENT

Having established that the shape of the measured anomaly in c_{11} at T_D is not seriously affected in the RF samples by either concentration inhomogeneity rounding or dynamic critical effects (with the possible exception of

TABLE II. Results of measurements of concentration inhomogeneities for samples with As concentrations x, showing total variation in: apparent birefringence [$\delta(\Delta n_{ca})$], arsenic concentration (δx), transition temperature (δT_D), and reduced temperature ($\delta\varepsilon$).

x	$\delta(\Delta n_{ca})$	δx	δT_D	$\delta\epsilon$
0	3×10^{-4}			
0.154 (10)[a]	5×10^{-4}	3×10^{-3}	0.15	2×10^{-2}
0.154 (10)[b]	11×10^{-4}	7×10^{-3}	0.3	4×10^{-2}
0.164 (10)	5×10^{-5}	3×10^{-4}	0.02	3×10^{-3}
0.984 (5)	7×10^{-5}	4.5×10^{-4}	0.06	7×10^{-3}

[a] [100] sample.

[b] [110] sample.

dynamic effects in the $x = 0.164$ sample), we were able to determine the static susceptibility critical exponent γ from our ultrasonic data as follows. Since in the static limit the soft-mode elastic constant $c_S = \frac{1}{2}(c_{11} - c_{12})$ must go to zero at $T = T_D$ for a second-order transition to occur, the "background" contribution $\frac{1}{2}(c_{11} + c_{12})$ is given by the minimum value of c_{11} at the transition, c_{11}^0, and can therefore be subtracted from the c_{11} data to obtain the behavior of c_S directly. This procedure requires that $\frac{1}{2}(c_{11} + c_{12})$ be independent of temperature in the critical region—a plausible assumption, since this elastic constant corresponds to a mode of the wrong symmetry to couple to the distortion.[21]

One test of the validity of the assumption that $c_S = 0$ at $T = T_D$ is the comparison of measurements of $c_{11} - c_{11}^0$ and c_S at temperatures as close to T_D as possible. In order to make such a comparison, we performed a direct measurement of c_S using shear waves propagating in the [110] direction in a sample prepared from the same piece of as-grown crystal as was the $x = 0.154$ [100] sample. Results for c_S in this sample, compared with the $c_{11} - c_{11}^0$ results for the same concentration, are presented in Fig. 2. The apparent convergence of the two curves as the temperature is reduced suggests that c_S does indeed vanish at $T = T_D$, although high attenuation in the [110] sample prevents this comparison from extending into the critical region. (The deviation at high temperatures is explicable in terms of the usual variation of elastic constants with temperature due to anharmonic effects.) We must also consider the possibility of an anomaly in $\frac{1}{2}(c_{11} + c_{12})$ near T_D, which may occur if there is a nonzero Jahn-Teller coupling to the corresponding (totally symmetric) A_{1g} mode (a dilation or contraction of the unit cell in the basal plane), since such a coupling is allowed by symmetry considerations.[30,31] Proper resolution of both these questions requires accurate measurements of $\frac{1}{2}(c_{11} + c_{12})$ right through T_D. This can be done by using longitudinal pulses propagating in the [110] direction to measure $\frac{1}{2}(c_{11} + c_{12}) + c_{66}$, then subtracting c_{66}, measured using shear waves propagating in the [100] direction.[20] Preliminary results of such measurements[29] for $x = 0.154$ show that $c_S = 0$ at $T = T_D$, within the rather large absolute experimental uncertainty of ±1 GPa in the difference between c_{11} and $\frac{1}{2}(c_{11} + c_{12})$. Attempts to detect an anomaly in $\frac{1}{2}(c_{11} + c_{12})$ have so far been inconclusive, owing to problems associated with a slight sample misalignment, although it does appear that if such an anomaly does exist it is too small to significantly affect the critical-exponent measurements.

The critical exponent γ was found from the slope of a log-log plot of $c_{11} - c_{11}^0$ versus reduced temperature ϵ for $\epsilon > 0$; the values used for T_D and c_{11}^0 were then adjusted, within the limits allowed by experimental uncertainty, to give the best possible straight line. Results for "best" values of c_{11}^0, T_D, and γ are shown in Fig. 3. The uncertainty in γ was determined by the amount it changed for different values of T_D and c_{11}^0 which still gave reasonable straight lines. Naturally on a log-log plot the experimental scatter of the data is enhanced at small ϵ, so the most heavily weighted data were generally in the range $10^{-2} < \epsilon < 10^{-1}$, above which ϵ is probably outside the critical region.

The measured exponent $\gamma = 1.15 \pm 0.05$ for $DyVO_4$ is greater than the classical value $\gamma = 1$,[32] which confirms earlier results[33] that $DyVO_4$ is not a good mean-field system.[34] It seems to be lower, however, than the nearest-neighbor Ising value[35] $\gamma = 1.25$ for spatial dimension $d = 3$ estimated from birefringence measurements on $DyVO_4$.[14]

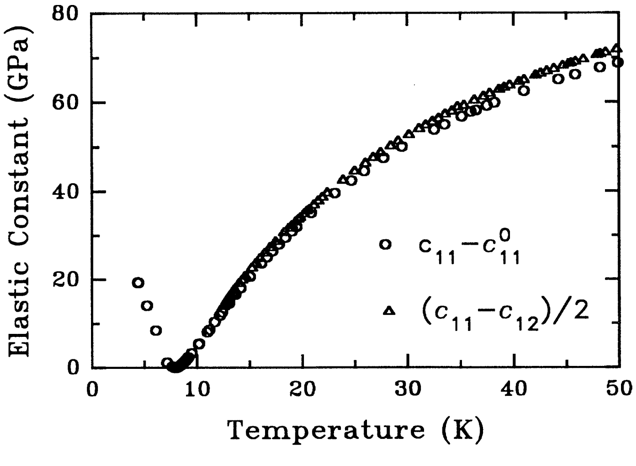

FIG. 2. Comparison of $\frac{1}{2}(c_{11} - c_{12})$ with $c_{11} - c_{11}^0$ for $x = 0.154$.

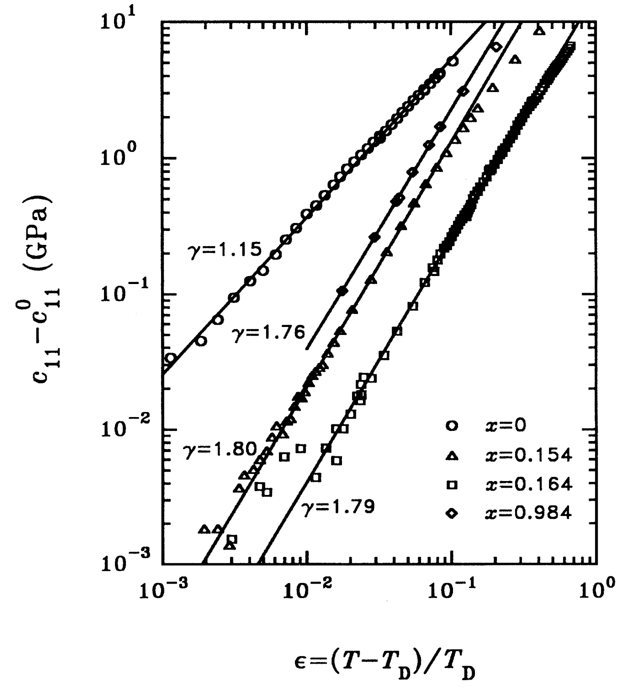

FIG. 3. Log-log plot of soft-mode elastic constant vs reduced temperature ϵ for all four [100] samples.

The measured critical exponents in random-field samples with $x = 0.154$, 0.164, and 0.984 are $\gamma = 1.80 \pm 0.06, 1.79 \pm 0.07$, and 1.76 ± 0.15 respectively, which are significantly higher than for the pure compound. The agreement between samples is well within experimental uncertainty, with the uncertainty for $x= 0.984$ being the highest owing to the small number of data points taken in the critical region. As noted above, the results for the strongest RF system, $x = 0.164$, are complicated by the possibility that dynamic rounding of the susceptibility may partially mask the static critical behavior and invalidate the background subtraction method used to extract γ from the data, so the value of γ obtained for this sample may not be reliable. The weighted average of the values for the other two samples gives $\gamma = 1.79 \pm 0.06$ for the RF Ising system $\mathrm{Dy(As}_x\mathrm{V}_{1-x})\mathrm{O}_4$.

This value for γ is compatible with the results of previous Monte Carlo simulations for the nearest-neighbor RFIM, which yielded $\gamma = 2.0 \pm 0.5$ (Ref. 36) and $\gamma = 1.7 \pm 0.2$ (Ref. 37). We also note that there is good agreement between our value of γ and the less precise value, $\gamma = 1.75 \pm 0.2$, obtained from the earlier neutron scattering measurements[5] on the DAF $\mathrm{Fe}_{0.6}\mathrm{Zn}_{0.4}\mathrm{F}_2$ (see Sec. I). This agreement between the two measured values of γ suggests that the line shape used to analyze the neutron scattering data apparently gives reliable results for the critical exponents despite the reservations expressed in Ref. 5. Thus our data also give indirect evidence that the neutron scattering value of the correlation-length exponent, $\nu = 1.0 \pm 0.15$, is probably also valid since it was obtained using the same line shape as the susceptibility exponent. This reinforces the interesting observation that our result for γ in $\mathrm{Dy(As}_x\mathrm{V}_{1-x})\mathrm{O}_4$ and the previous results in DAF's are all consistent with the values $\gamma = \frac{7}{4}$, $\alpha = 0$, and $\nu = 1$ for the pure $d = 2$ nearest-neighbor Ising model,[38] i.e., consistent with a reduction in effective dimensionality from three to two in the presence of random fields, as suggested for the RFIM by Aharony, Imry, and Ma[39] and later predicted by Shapir,[40] although we recognize the likelihood that there is no exact dimensional reduction for the RFIM.[41,42] One theoretical model that raises doubts about this dimensional reduction hypothesis is the scaling theory for the RFIM critical exponents in $2 + \epsilon$ dimensions developed by Bray and Moore,[42] who found $\gamma = \frac{1}{2} + \frac{1}{\epsilon} = 1.5$ for $d = 3$; while their model predicts an increase in γ in the presence of a RF (the same trend that we observe experimentally), their value is smaller than our result, falling well outside the range of our experimental uncertainty. It is interesting to note that our results for γ are not in agreement with a renormalization-group calculation by Toh[43] for the mean-field RFIM, which yields the much lower value $\gamma = 1.2$. Since our measurements in pure DyVO_4 are performed in a range of reduced temperatures where short-range interactions lead to nonclassical exponents, it is not surprising that our result for the mixed RF system also yields a value for γ that differs from predictions of mean-field theory and is closer to predictions for the nearest-neighbor RFIM.

IV. EFFECT OF CONCENTRATION INHOMOGENEITY

In order to study the effects of the measured variations in x on our critical-exponent measurements, numerical simulations have been performed, using essentially the method of KBFJ. We assumed a form $c(T,T_D)$ for the "soft-mode" elastic constant $c_S = c_{11} - c_{11}^0$, and integrated over a distribution of T_D's in the sample to get what should be the measured elastic constant $c_M(T)$. For simplicity, we assumed a uniform distribution of T_D's, restricted to the range $T_a < T_D < T_b$. Our birefringence measurements suggest that this is at least a reasonable approximation, and KBFJ suggest that the smearing of a transition should not be particularly sensitive to the precise form of this distribution function, anyway. We used the following functional form for the elastic constant:

$$c(T,T_D) = \begin{cases} G\epsilon^{\gamma}, & T > T_D \\ G'(-\epsilon)^{\gamma'}, & T < T_D \end{cases} \tag{1}$$

where G and G' are the critical coefficients. Since our measured values for $c_{11} - c_{11}^0$ will include a contribution due to sample dilation when $T < T_D$ we should perhaps include a term proportional to $(-\epsilon)^{\beta}$ (where β is the order parameter exponent, and the order parameter is proportional to the lattice distortion); however, we do not have a good idea what either β or the coefficient should be, and we are mostly interested in the region $T > T_b$ anyway, in which $T > T_D$ over the whole sample. Our G' and γ' are merely effective coefficients used to describe the data.

The observed elastic constant, obtained by averaging Eq. (1) over all T_D's, will be[25,44]

$$c_M(T) = \frac{1}{T_b - T_a} \int_{T_a}^{T_b} dT_D \; c(T,T_D)$$

$$= [\delta T_D (1+\gamma)(\bar{T}_D^{\gamma})]^{-1} \times \begin{cases} G[(T-T_a)^{1+\gamma} - (T-T_b)^{1+\gamma}], & T > T_b & \text{(2a)} \\ G(T-T_a)^{1+\gamma} + G'(T_b - T)^{1+\gamma}, & T_a < T < T_b & \text{(2b)} \\ G'\left[(T_b - T)^{1+\gamma} - (T_a - T)^{1+\gamma}\right], & T < T_a & \text{(2c)} \end{cases}$$

where $\delta T_D = T_b - T_a$, $\bar{T}_D = \frac{1}{2}(T_a + T_b)$, $\delta T_D \ll \bar{T}_D$, and we have assumed $\gamma = \gamma'$. Equation (2a) determines the behavior over most of the region of interest to us, but it is Eq. (2b) which determines the temperature T^0 and value c_M^0 of the minimum. T^0 is determined largely by the ratio G/G', i.e., by the asymmetry of the minimum, and c_M^0 is determined mostly by the magnitude of δT_D (we see immediately that $c_M^0 \neq 0$ for $\delta T_D \neq 0$). Furthermore, the larger the exponent under investigation, the smoother is the minimum, and the less effect a given small concentration gradient will have on the critical behavior. We therefore expect to see a smaller effect in our measurements, for which $\gamma \approx 1.8$, than in the specific-heat experiments discussed by Belanger *et al.*,[25] for which the exponent is $\alpha \cong 0$, or in the neutron scattering experiments which they discuss, for which the correlation-length exponent is $\nu \cong 0.6$.

In our analysis, we concentrated on the $x = 0.154$ sample, since it has the greatest $\delta\epsilon = \delta T_D / T_D$ (Table II). We chose what appeared from our measurements to be reasonable values for the parameters G, G', and γ, calculated $c_M(T)$ from Eqs. (2), then proceeded to analyze the results exactly as we had analyzed our experimental data to determine γ.

Figure 4 shows the simulation results for $\gamma = 1.80$, $\bar{T}_D = 7.9480$ K, $\delta T_D = 0.15$ K, $G = 84$ GPa, and $G' = 95$ GPa, compared with the result for a perfectly homogeneous sample ($\delta T_D = 0$). The inhomogeneities cause the transition to be more rounded, with the minimum shifted to $T^0 = 7.9509$ K (a shift of 4×10^{-4} in ϵ) and raised from zero to $c_M^0 = 7.2 \times 10^{-3}$ GPa. These data are compared on a log-log plot in Fig. 5, which also shows the simulated data as we would have plotted them in a first attempt at finding γ, i.e., $c_M(T) - c_M^0$ versus $(T - T^0)/T^0$. Evidently, the difference between the three curves is too small to be resolved (within the precision of our ultrasonics experiments) on such a plot for $\epsilon > 2 \times 10^{-2}$, which is approximately twice the upper limit of transition temperatures in the sample.

In our ultrasonics experiments there is experimental uncertainty associated with the measurement of c_M^0 and T^0, so they become adjustable parameters in the determination of γ. Figure 6 shows the simulated data with offsets $T^{0'}$ and $c_M^{0'}$ adjusted for a best straight line in the region $10^{-2} < \epsilon < 10^{-1}$, as was done for our critical plots (Fig. 3). The parameters which give the best fit are: $T^{0'} = 7.9538$ K, $c_M^{0'} = 6.50 \times 10^{-3}$ GPa, and $\gamma = 1.787$.

The results of our simulation suggest that the uncertainty in γ due to concentration inhomogeneities is significantly less than the overall uncertainty in γ, even for a fairly large spread in transition temperature. For $\delta\epsilon = 0.02$, the "true" value of γ is greater than the value determined by our method, by 0.013; our lowest experimental uncertainty in γ was ± 0.06. For this value of $\delta\epsilon$, the region of serious "rounding" for $\epsilon > 0$ is restricted

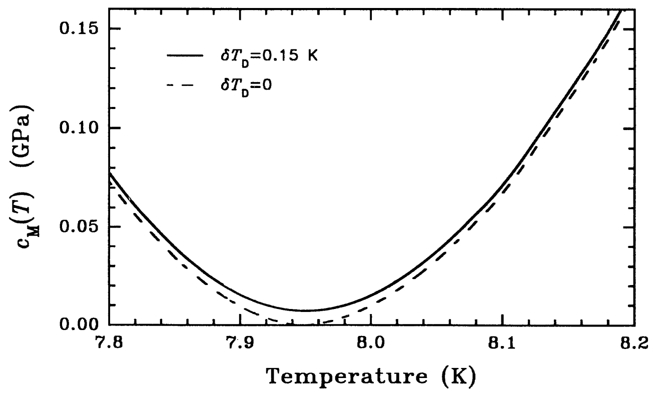

FIG. 4. Simulated results for smearing, due to concentration inhomogeneities, of the phase transition in the $x = 0.154$ sample, for $\delta T_D = 0.15$ K, compared with the case for a uniform ($\delta T_D = 0$) sample.

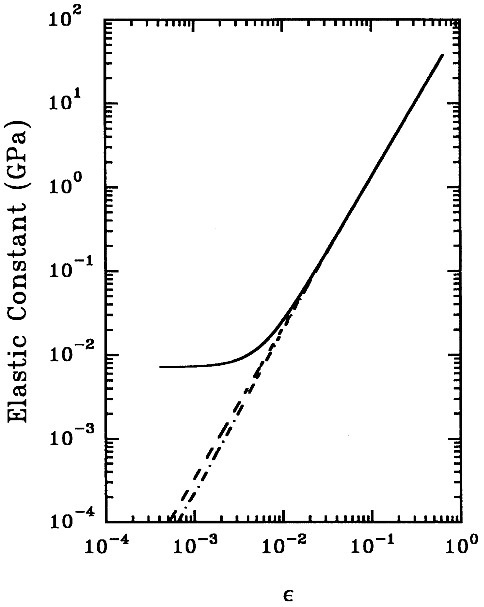

FIG. 5. Log-log plot of simulated data for $c_M(T)$ vs $(T - \bar{T}_D)/\bar{T}_D$, for $\delta T_D = 0$ (– –), and $\delta T_D = 0.15$ K (—), along with a plot of $c_M(T) - c_M^0(T)$ vs $(T - T_0)/T_0$ (- ·).

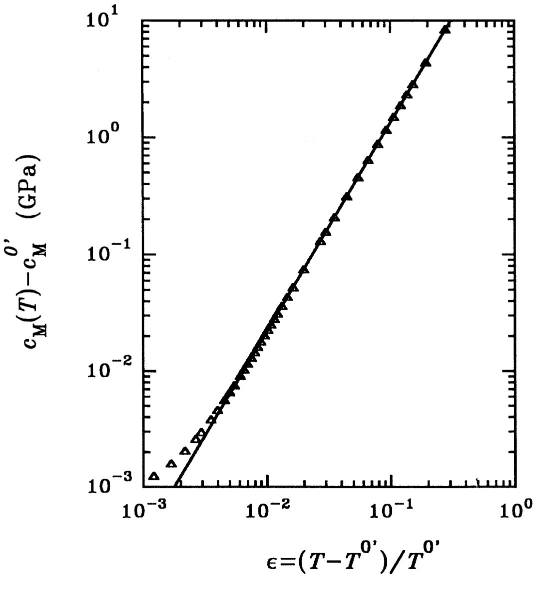

FIG. 6. Simulation data and fit, using the same method as for the experimental data (Fig. 3), for best fit in the range $0.01 < \epsilon < 0.1$.

to $\epsilon < \delta\epsilon$ (Fig. 5). The mean transition temperature $\bar{T}_D$ is less than the value determined by our method, by $T^{0'} - \bar{T}_D = 0.0058$ K, as compared with the experimental uncertainty, ± 0.01 K, in the temperature of the minimum in c_{11}.

For this simulation we have deliberately chosen parameters corresponding to the sample with the greatest measured inhomogeneities. The effect of concentration inhomogeneities on the measurement of γ in the $x = 0.164$ sample, for which $\delta\epsilon$ is lower by an order of magnitude, is negligible.

V. CONCLUSIONS

We have successfully used ultrasonic velocity measurements to determine the susceptibility critical exponent γ in the random-field Ising system $\mathrm{Dy(As}_x\mathrm{V}_{1-x})\mathrm{O}_4$. We find that the random-field susceptibility exponent, $\gamma = 1.79 \pm 0.07$,[45] is substantially higher than in the pure system, in which $\gamma = 1.15 \pm 0.05$. In the range of reduced temperature over which the critical-exponent measurements could be performed ($10^{-3} < \epsilon < 10^{-1}$), the observed power-law behavior is determined by short-range Ising interactions in the presence of static random fields, so that our results are potentially important for helping to elucidate the critical properties of the nearest-neighbor RFIM. Our principal finding, that γ is considerably modified by random fields, can be taken as clear evidence that the RFIM belongs to a new universality class.

Birefringence measurements on our samples, combined with numerical simulations, have shown that the values determined for γ are not significantly affected by concentration inhomogeneities even in the $x = 0.154$ sample, which had the greatest variation in As concentration. The effect of a given spread $\delta\epsilon$ in reduced transition temperature on our determination of γ is smaller than that for the critical-exponent analysis discussed by KBFJ (Refs. 24 and 25) because (a) the exponent under consideration here is larger, so inhomogeneities cause merely a further rounding of a smooth transition, which is considerably less dramatic than the rounding of, say, a sharp cusp, and (b) the transition under consideration here is more symmetric (our $G/G' \sim 0.9$, compared with their $A/A' \approx 1.6$ for the specific heat and $\kappa_0^+/\kappa_0^- \approx 0.7$ for the correlation length), so the "apparent" transition temperature is not much different from the "true" transition temperature.

Future work will focus on improving the accuracy of the elastic-constant measurements for c_{11} and $\frac{1}{2}(c_{11}+c_{12})$ so that the soft-mode elastic constant $c_S = \frac{1}{2}(c_{11} - c_{12})$ can be measured on an *absolute* scale near T_D. Further measurements are also planned as a function of frequency to make a more quantitative study of dynamic effects in our strongest random-field sample of $\mathrm{Dy(As}_x\mathrm{V}_{1-x})\mathrm{O}_4$.

ACKNOWLEDGMENTS

The crystals used in these experiments were provided by B. E. Watts and the Crystal-Growing Group at the Clarendon Laboratory, Oxford. Helpful discussions with R. J. Gooding and B. W. Southern were much appreciated. Some experimental assistance was provided by M. C. Maliepaard and H. M. Elmehdi. Research support was provided by the Natural Sciences and Engineering Research Council of Canada, by Queen's University, and by the University of Manitoba.

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[1] Y. Imry and S.-k. Ma, Phys. Rev. Lett. **35**, 1399 (1975).

[2] For recent reviews, see R.A. Cowley, in *Neutron Scattering* (Part C), Vol. 23 of *Methods of Experimental Physics*, edited by K. Sköld and D.L. Price (Academic, Orlando, 1986), p.1; V. Jaccarino and A.R. King, Physica A **163**, 291 (1990).

[3] S. Fishman and A. Aharony, J. Phys. C **12**, L729 (1979); J.L. Cardy, Phys. Rev. B **29**, 505 (1984).

[4] D.P. Belanger, A.R. King, V. Jaccarino, and J.L. Cardy, Phys. Rev. B **28**, 2522 (1983).

[5] D.P. Belanger, A.R. King, and V. Jaccarino, Phys. Rev. B **31**, 4538 (1985).

[6] D.P. Belanger, V. Jaccarino, A.R. King, and R.M. Nicklow, Phys. Rev. Lett. **59**, 930 (1987); V. Jaccarino and A.R. King, J. Phys. (Paris) Colloq. **49**, C8-1209 (1988).

[7] J.H. Page, D.R. Taylor, and S.R.P. Smith, J. Phys. C **17**, 51 (1984).

[8] For a review see G.A. Gehring and K.A. Gehring, Rep. Prog. Phys. **38**, 1 (1975).

[9] D.R. Taylor, J.T. Graham, J.D. MacArthur, R.D. Heyding, B.A. Judd, J.H. Page, G.H. Pelletier, and B.E. Watts, J. Phys. Chem. Solids **51**, 197 (1990).

[10] A. Kasten, Z. Phys. B **38**, 65 (1980).

[11] G.A. Gehring, S.J. Swithenby, and M.R. Wells, Solid State Commun. **18**, 31 (1976).

[12] D.R. Taylor, E. Zwartz, J.H. Page, and B.E. Watts, J. Magn. Magn. Mater. **54-57**, 57 (1986).

[13] We did not perform ultrasonics measurements on pure $\mathrm{DyAsO_4}$, in which the transition is *first* order.

[14] G.A. Gehring, R.T. Harley, and R.M. Macfarlane, J. Phys. C **13**, 3161 (1980).

[15] D.A. Huse and C.L. Henley, Phys. Rev. Lett. **54**, 2708 (1985).

[16] J.T. Graham, D.R. Taylor, D.R. Noakes, and W.J.L. Buyers, Phys. Rev. B **43**, 3778 (1991); D.R. Taylor, J.T. Graham, T.P. Matthews, D.R. Noakes, and W.J.L. Buyers, J. Phys. (Paris) Colloq. **49**, C8-1239 (1988).

[17] J.T. Graham, M. Maliepaard, J.H. Page, S.R.P. Smith, and D.R. Taylor, Phys. Rev. B **35**, 2098 (1987).

[18] J.H. Page and J.T. Graham, J. Phys. (Paris) Colloq. **49**, C8-1227 (1988).

[19] R.L. Melcher and B.A. Scott, Phys. Rev. Lett. **28**, 607 (1972).

[20] J.R. Neighbours and G.E. Schacher, J. Appl. Phys. **38**, 5366 (1967).

[21] A similar technique has been used previously for $\mathrm{TmVO_4}$: J.H. Page and H.M. Rosenberg, J. Phys. C **10**, 1817 (1977).

[22] For an introduction to ultrasonic interferometry, see M.A. Breazeale, John H. Cantrell, Jr., and Joseph S. Heyman, in *Ultrasonics*, Vol. 19 of *Methods of Experimental Physics*, edited by Peter D. Edmonds (Academic, New York, 1981), p.67; for details of these particular measurements, see: J.T. Graham, Ph.D. thesis, Queen's University, Kingston, Canada, 1990.

[23] No indium layer was used in the absolute velocity measurements.

[24] A.R. King, I.B. Ferreira, V. Jaccarino, and D.P. Belanger, Phys. Rev. B **37**, 219 (1988).

[25] D.P. Belanger, A.R. King, I.B. Ferreira, and V. Jaccarino, Phys. Rev. B **37**, 226 (1988).

[26] The term "apparent birefringence" is used in recognition of the possible contribution from sample-thickness variations; see Refs. 9 and 24.

[27] J. Villain, J. Phys. (Paris) **46**, 1843 (1985); D.S. Fisher, Phys. Rev. Lett. **56**, 416 (1986); Jaccarino and King, Ref. 2.

[28] A.R. King, J.A. Mydosh, and V. Jaccarino, Phys. Rev. Lett. **56**, 2525 (1986); A.E. Nash, A.R. King, and V. Jaccarino, Phys. Rev. B **43**, 1272 (1991).

[29] H.M. Elmehdi, J. H. Page, and J. T. Graham (unpublished).

[30] R.J. Elliott, R.T. Harley, W. Hayes, and S.R.P. Smith, Proc. R. Soc. London, Ser. A **328**, 217 (1972).

[31] A similar A_{1g}-mode anomaly in c_{33} may have been observed previously in DyVO$_4$: G. Gorodetsky, B. Lüthi, and B.M. Wanklyn, Solid State Commun. **9**, 2157 (1971). We note that coupling to a totally symmetric mode can be the *dominant* Jahn-Teller interaction in certain cases, as has been observed in cerium ethyl sulfate; see J.T. Graham and J.H. Page, in *Phonon Scattering in Condensed Matter, Proceedings of the Fourth International Conference*, edited by W. Eisenmenger, K. Lassmann, and D. Dottinger (Springer-Verlag, Berlin, 1984), p. 278.

[32] See, for example, H.E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford University Press, Oxford, England, 1971).

[33] See Ref. 7, and references cited therein.

[34] Unlike, e.g., the Jahn-Teller systems TmVO$_4$ (see Ref. 21) and TbVO$_4$ [R.T. Harley and R.M. Macfarlane, J. Phys. C **8**, L451 (1975)].

[35] See, for example, J.C. Le Guillou and J. Zinn-Justin, Phys. Rev. B **21**, 3976 (1980), and references cited therein.

[36] A.T. Ogielski and D.A. Huse, Phys. Rev. Lett. **56**, 1298 (1986).

[37] A.P. Young and M. Nauenberg, Phys. Rev. Lett. **54**, 2429 (1985). We note that their prediction of a first-order transition is *not* supported by our results.

[38] L. Onsager, Phys. Rev. **65**, 117 (1944).

[39] A. Aharony, Y. Imry, and S.-k. Ma, Phys. Rev. Lett. **37**, 1364 (1976).

[40] Y. Shapir, Phys. Rev. Lett. **54**, 154 (1985).

[41] D.S. Fisher, Phys. Rev. Lett. **56**, 1964 (1986).

[42] A.J. Bray and M.A. Moore, J. Phys. C **18**, L927 (1985).

[43] H.S. Toh (unpublished).

[44] We note that in Eqs. (5a)–(5c) of Ref. 25 $\bar{T}_c$ should be replaced by $(\bar{T}_c)^{-\alpha}$.

[45] This uncertainty includes a small contribution due to inhomogeneity in x.