Measurement of static critical exponents for a structural Ising-model phase transition with random strains: Dy(As$_x$V$_{1-x}$)O$_4$

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The critical exponents β, δ, and γ have been measured for the tetragonal-orthorhombic phase transition in Dy(As$_x$V$_{1-x}$)O$_4$. For pure DyVO$_4$ the exponents are close to Ising $d = 3$ values. For $x = 0.05$ and $x = 0.154$ the random strain fields arising from As-V substitutions depress the transition temperature and alter the critical behavior. The exponents β, δ, and γ for the two mixed samples agree well with each other: γ increases to 1.55 compared to 1.15 for pure DyVO$_4$, and δ increases to 5.8 compared to 4.1 for DyVO$_4$. However, β remains unchanged near 0.33 within experimental error. Although the increase in γ is consistent with exponents expected for the random-field Ising model, the result for β is not. The measured exponents satisfy the scaling relation $\gamma = \beta(\delta - 1)$ within the experimental uncertainties, and together with another scaling relation, $\alpha + 2\beta + \gamma = 2$, allow us to infer that the critical specific-heat behavior changes from divergent ($\alpha > 0$) in pure sample to cusplike ($\alpha < 0$) in the mixed samples.

I. INTRODUCTION

There is still little understanding of the critical properties of random-field Ising-model (RFIM) systems in spite of efforts continuing over many years. Recent review articles by Belanger and Young,[1] and by Belanger[2] describe the status of theoretical, experimental, and numerical investigations of the RFIM phase transition. Although the RFIM is a relatively simple system in which the effects of randomness can be investigated, theoretical analysis still encounters such difficulties that important questions cannot be resolved. Studies of dilute antiferromagnetic (DAF) systems have provided valuable experimental information on the RFIM, but the combination of randomness and dilution introduces such long equilibration times that reliable results for critical exponents are hard to obtain. The same factors also stand in the way of convergence of Monte Carlo simulations so that numerical estimates of exponents contain large uncertainties even after extensive computational effort. It is accepted[1] that the universality class of the RFIM differs from that of the pure Ising model, but the available critical exponent data have not been sufficient to characterize the new class.

In recent years a structural realization of the RFIM has been proposed,[3] which has the prospect of providing new experimental information on RFIM properties. It corresponds closely to the type of system in which Imry and Ma[4] first suggested that RFIM effects might be observed. The pure compounds DyAsO$_4$ and DyVO$_4$ undergo tetragonal-orthorhombic phase transitions that are well described by Ising models.[5] In the mixed crystals Dy(As$_x$V$_{1-x}$)O$_4$ the strain fields generated by the Dy-As size mismatch play the same role as random magnetic fields in the RFIM. In contrast to the DAF systems, the order parameter is a uniform mode in these systems and can be measured by bulk probes.

Measurements of the susceptibility exponent γ were reported by Graham, Page, and Taylor[6] for Dy(As$_x$V$_{1-x}$)O$_4$. From ultrasonic elastic constant results they found $\gamma = 1.79 \pm 0.07$ as an average for mixed samples with several compositions, significantly larger than their result $\gamma = 1.15 \pm 0.05$ for pure DyVO$_4$ ($x = 0$). Optical birefringence measurements by Taylor and Reza[7] on a $x = 0.154$ sample also gave an increased value for γ, namely, 1.57 ± 0.15, but the same paper reported order-parameter measurements that indicated that the order-parameter exponent β remained unchanged at $\beta = 0.31 \pm 0.05$. The latter result is very surprising because a small value of β is expected for the RFIM based on results, albeit inconclusive, from theoretical,[8] numerical,[9] and experimental[10] investigations. We have therefore repeated and extended our previous experiments[7] to check β and to more fully characterize the set of static critical exponents for this system. In this paper we report results for mixed crystals of two compositions as well as pure DyVO$_4$, and give results for β obtained by two independent methods. In addition we report the first measurement of the critical "isotherm" exponent δ. By incorporating measurements of γ carried out at the same time we are thus able to test the scaling relation

$$\gamma = \beta(\delta - 1) \tag{1}$$

for the static critical exponents.

A. The Dy(As$_x$V$_{1-x}$)O$_4$ phase transition

There have been many studies of the phase transitions in DyAsO$_4$, DyVO$_4$, and Dy(As$_x$V$_{1-x}$)O$_4$. The early work on DyVO$_4$, summarized in the review article by Gehring and Gehring,[5] identified the origin of the phase transition as a coupling between nearly degenerate Dy electronic levels and lattice distortions. Tetragonal-orthorhombic transitions occur near 11 K and 14 K for

DyAsO$_4$ and DyVO$_4$, respectively. For these systems the effective ion-ion interaction can be described by a pseudospin Ising Hamiltonian to an excellent approximation.[5] The pseudospin directions correspond to the two orientations of the orthorhombic unit-cell axes. The order parameter is proportional to the B_{1g} strain that develops below the transition temperature T_D.

The critical properties of this and related Jahn-Teller systems are expected to be classical because there is a coupling of the ions to uniform strains.[11] However, for Dy compounds this coupling is apparently small compared with short-range interactions: the temperature regime for classical behavior is consequently too small to be accessible experimentally, and the observed power-law exponents are close to $d = 3$ Ising values.[12,13]

In the mixed crystals Dy(As$_x$V$_{1-x}$)O$_4$, the As-V substitutions generate random, static strain fields, one component of which has the right symmetry to couple to the order parameter. The RFIM Hamiltonian should then apply:

$$\mathcal{H} = -\sum_{ij} J_{ij}\sigma_i\sigma_j - \sum_i h_i\sigma_i, \tag{2}$$

where h_i is expected to have approximately a Gaussian distribution about $h = 0$. The interaction parameters J_{ij} are of similar magnitudes in DyAsO$_4$ and DyVO$_4$, and therefore we do not expect random exchange behavior in the mixed compounds. For x near 0 or 1, well-defined tetragonal-orthorhombic transitions are observed at reduced transition temperatures T_D, while for intermediate values of x the phase transition is suppressed.

An important feature of experiments in this system, and an essential one for determining the critical exponents γ and δ, is that an external ordering field is easily applied. This could have the form of a stress parallel to a basal plane axis (x or y) to favor one of the orientations of the orthorhombic axes. In practice, it is more convenient to apply a magnetic field, say B_x, which induces a corresponding stress through coupling between the Dy magnetic moment and unit-cell distortions. An expansion of the free energy (for example see Page et al.[14]) implies that the ordering field is proportional to B_x^2/T. This point was considered carefully by Gehring, Harley, and Macfarlane[15] who concluded that B_x^2 was a valid ordering field, at least near T_D. We expect that over a wider temperature range B_x^2/T may be taken as an ordering field to high accuracy.

B. Experimental procedure

Flux-grown Dy(As$_x$V$_{1-x}$)O$_4$ crystals with $x =$0, 0.050, and 0.154 were obtained from the Clarendon Laboratory, Oxford. Details of their characterization are given in a previous publication.[16] Of particular interest is the observation that composition inhomogeneities are small and unlikely to affect the determination of critical exponents.[6,16] The crystals were cut and polished with plane faces perpendicular to the c axis separated by approximately 1 mm. They were selected for good optical quality by examination of their conoscopic interference figures with a polarizing microscope at room tempera-

ture. They were mounted in a strain-free manner on a sample holder in a He flow cryostat so that the crystal c axis was horizontal and a axis vertical. The sample holder could be rotated about the vertical axis. For optical birefringence measurements a beam of linearly polarized light (He-Ne laser, 633 nm) was transmitted through the sample with polarization axis at 45° to the a axis as shown schematically in Fig. 1. A beam expander was mounted in front of the laser for shaping and smoothing the beam. A photoelastic crystal modulated the incident beam at 50 kHz. After traversing the sample where it becomes elliptically polarized in the distorted (orthorhombic) phase, the beam passed through an analyzer set perpendicular to the incident polarization axis and was detected by a photomultiplier. The output signal was fed to a lock-in amplifier driven at the modulator frequency. A Soleil-Babinet compensator placed between the sample and the analyzer was used to cancel the phase retardation in the crystal as indicated by a null signal from the lock in. The sample retardation ϕ was determined as a function of temperature and magnetic field by manual setting of the compensator. The sample birefringence $\Delta n = n_a - n_b$ was obtained from the relation $\phi = \Delta n(2\pi/\lambda)l$, where l is the sample thickness and λ the wavelength of the incident light. Δn should be accurately proportional to the order parameter a-b in the orthorhombic phase.[15] Above T_D an applied ordering field induces birefringence in proportion to the susceptibility χ of the system. To determine $\chi(T)$ we measured $\Delta n(T)$ for a series of fields B_x up to 170 mT and carried out fits of Δn versus B_x^2. Good linear fits were obtained except for temperatures very close to T_D.

It is well known[5] that DyVO$_4$ and related crystals are twinned in the orthorhombic phase, with domain walls parallel to {110} axes separated by a few μm. The twinned domains have opposite orientations of the orthorhombic axes, and hence the birefringence in the ordered phase largely cancels out when sampled over the area of the laser beam. In order to obtain $\Delta n(T)$ we measured the birefringence in applied magnetic fields up to 170 mT and extrapolated Δn data taken above about 70 mT, where the sample was in a single-domain state, to zero field. The order-parameter exponent β was then determined from the zero-field values. In these measurements it is important to test for the presence of hysteresis, which if present would imply that we were not measuring equilibrium properties. Measurements in DAF systems are severely limited by long equilibration times near and below the transition temperature.[1] In our procedure we systematically measured Δn both for increasing and for decreasing fields at each temperature, and compared the results carefully. In contrast to the DAF systems, there was, in general, no observable hysteresis on the
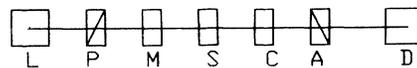
FIG. 1. Schematic illustration of the birefringence apparatus. Components are L = light source, P = polarizer, M = modulator, S = sample, C = compensator, A = analyzer, and D = detector.

time scale of our measurements (100 s) throughout the critical regime. As the temperature was reduced below about 0.9 T_D, some hysteresis appeared, but only in the twinning-detwinning regime. At the very lowest temperatures ($T < 0.5T_D$) hysteresis could also be observed in the single domain state.[17]

As a check of our results for β, we also measured the order parameter in the twinned phase, with no applied field, by an entirely different technique. The method is based on the diffraction of light from the twinned domains.[18] The domain-wall separations are comparable in magnitude to the wavelength of light, and sufficiently uniform to allow the observation of Bragg diffraction at scattering angles of a few degrees. The intensities of the Bragg peak were recorded as a function of temperature. According to arguments made previously for a similar situation,[19] the scattering strength is a quadratic function of the order parameter. Thus near T_D the peak intensity is proportional to $t^{2\beta}$, where t is the reduced temperature $(T_D - T)/T_D$. These measurements can be carried out during the same run as the birefringence experiments by rotating the sample c axis a few degrees away from alignment with the laser beam and locating the diffracted beam on a screen. The diffracted peaks were generally rather broad, presumably because of a distribution of domain widths.[18] Rather than attempting to measure the integrated intensity of the scattered beam, we adjusted the aperture of a photomultiplier detector to monitor the intensity of the peak. Preliminary results for pure and mixed samples were recently reported.[18]

The dependence of the order parameter on ordering field exactly at the transition temperature defines the critical exponent δ through the relation $\Delta n \propto (B_x^2/T_D)^{1/\delta}$. It was straightforward to extract this relationship from our measurements of Δn as a function of temperature and field, although there is some uncertainty in identifying the transition temperature accurately.

II. EXPERIMENTAL RESULTS

The emphasis in these experiments was to get good data for the $x = 0.05$ sample. Measurements on the pure $DyVO_4$ sample were less extensive, since they were intended as a comparison check. They did not reach as far into the critical regime, perhaps because the $DyVO_4$ sample was quite small and more difficult to mount without strain. Most of the data for the $x = 0.154$ sample were reported previously.[7]

Critical exponents were extracted from the data by means of power-law fits with both the exponent and the transition temperature T_D as variables. In the fitting procedure, T_D was allowed to vary between experiments, and even during the same run because changes in T_D were apparent as a result of the strains in mounting the sample, and in unavoidable temperature gradients in the sample holder. This should not affect the accuracy of the critical exponents. The quoted uncertainties in the exponents were estimated from the variations in the parameters when fits were made over different ranges of reduced temperature in the critical regime. While the quality of fit to some of the data was not high, the con-
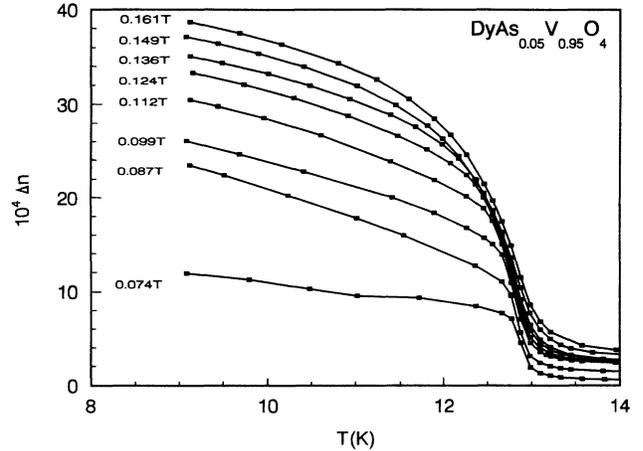
FIG. 2. A set of data for birefringence Δn vs temperature for a range of magnetic fields. For small fields the sample is not a single domain.

sistency between repeated measurements and among the overall set of experiments gives confidence in the results.

A. Order parameter exponent β

A set of data for birefringence, Δn, as a function of temperature and field for the $x = 0.05$ sample is shown in Fig. 2. Below T_D the sample is not a single domain for fields less than 90 mT. Figure 3 shows $\Delta n(T)$ results obtained by extrapolating the data of Fig. 2 to zero field. The uncertainties in the extrapolation are indicated by the error bars. Similar data for all three samples were fitted to a power law of the form $\Delta n \propto t^\beta$, with the results given in Table I. Log-log plots for the same samples are shown in Fig. 4. For pure $DyVO_4$ our result β=0.35± 0.03 is consistent with $d = 3$ Ising exponents and with previous measurements.[12] However, it is unexpected that
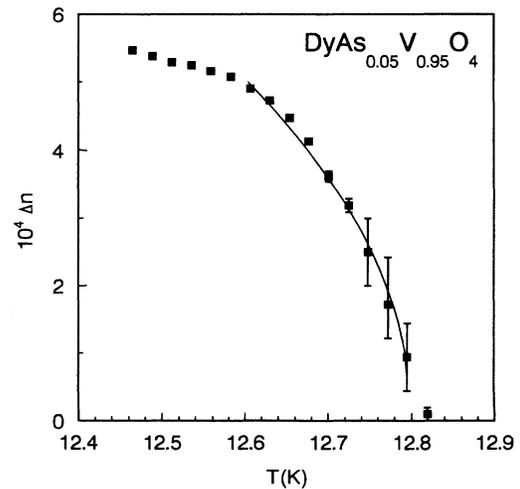
FIG. 3. Birefringence Δn vs temperature for the sample $x = 0.05$. Δn was obtained by extrapolating data taken for a single-domain sample in an ordering field to zero field. The solid line is a power-law fit with exponent $\beta = 0.32$.

TABLE I. Measured critical exponents for $Dy(As_xV_{1-x})O_4$.

x	T_D (K)	β (Δn)	β (scatt.)	δ	γ
0	13.5	0.35(3)	0.32(3)	4.1(5)	1.15(5)
0.050	12.8	0.32(3)	0.30(3)	5.5(5)	1.52(6)
0.154	7.8	0.34(3)	0.31(3)	6.1(7)	1.57(6)

β does not change, within experimental error, for the mixed samples even though T_D is sharply reduced.

The results of the Bragg-scattering experiments were analyzed by fitting the Bragg-peak intensities at temperatures close to T_D to the expected relation $t^{2\beta}$. This procedure is only valid if the diffraction line shapes and the configuration of domains is independent of temperature. From our observations this appeared to be true to a good approximation. Figure 5 shows log-log plots of these data and Table I gives the values of β determined from the slopes. There is clearly excellent agreement with the values for β from the birefringence results.

B. Critical isotherm exponent δ

The birefringence (order parameter) as a function of ordering field at various fixed temperatures is shown in Fig. 6 for the $x = 0.05$ sample. These isotherms show distinct behaviors above and below the transition temperature. Above the transition temperature $(T > T_D)$ Δn as a function of ordering field is almost linear. Below T_D the behavior becomes quite nonlinear, although the initial dependence (up to fields of about 70 mT) appears to be linear close to T_D. This linear regime is interpreted as the signature of progressive detwinning of the sample with increasing fields.[17] The Δn data in the single-domain regime were fitted to the power law $(B_x^2/T_D)^{1/\delta}$. The values of δ determined for the three samples in this way are given in Table I. The experimental uncertainties

FIG. 5. Log-log plots of diffracted beam intensity (arbitrary units) vs reduced temperature for the three Dy compounds, approximately 0.32 in each case. For $TbVO_4$ the slope gives $\beta = 0.47$.

are rather large because of the difficulty of locating T_D and the necessity to discard the low-field data because of incipient twinning. Log-log plots showing the fits are given in Fig. 7 and indicate that δ is significantly larger for the mixed samples than for pure $DyVO_4$.

C. Susceptibility exponent γ

As mentioned above, the order parameter Δn is a linear function of ordering field B_x^2/T, at least above T_D. The susceptibility χ, determined from the the slope as a function of temperature, was fitted to the relation $\chi^{-1} \propto |t|^{\gamma}$. The results for the same three samples are shown in Table I, and are illustrated in the form of log-log plots in Fig. 8. For pure $DyVO_4$ our result $\gamma = 1.15 \pm 0.05$ is intermediate between the values 1.0

FIG. 4. Log-log plots for order parameter (Δn) and reduced temperature for the three samples. Table I gives values for β for each sample, equal within experimental error.
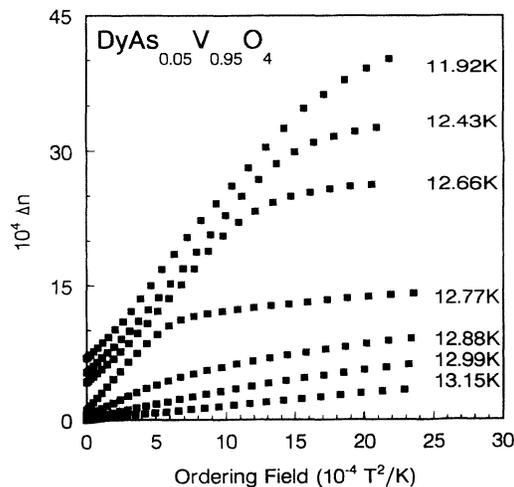
FIG. 6. Birefringence for the $x = 0.05$ sample vs ordering field for temperatures near $T_D = 12.8$ K. The ordering field (see text) is B_x^2/T.
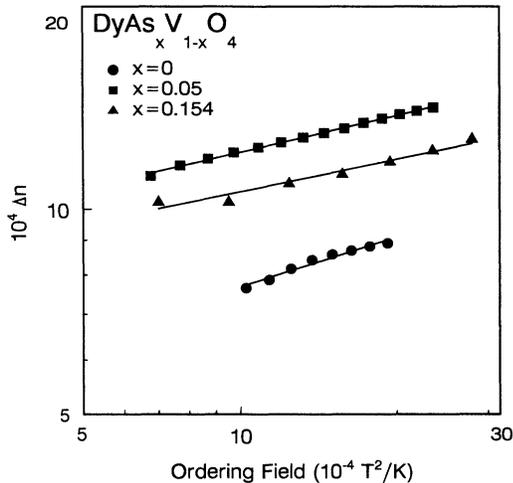
FIG. 7. Log-log plots of Δn vs ordering field at T_D for the three samples. The slopes give δ as given in Table I.

for a mean-field system and 1.25 for a nearest-neighbor Ising system. Our data were not of sufficient quality to allow us to attempt to find a crossover from one regime to the other. For the mixed samples, the values 1.52 and 1.57 for γ are clearly larger than that for pure DyVO$_4$.

III. DISCUSSION

The results of these experiments may be summarized as follows: (a) our values of the exponents β, δ, and γ for pure DyVO$_4$ are consistent with previous measurements as far as they are available and reasonably consistent with expected $d = 3$ Ising exponents; (b) the exponents β, δ, and γ for the two mixed samples agree well with each other, but are not the same as those for pure DyVO$_4$; (c) while δ and γ change on going from the pure to the mixed system, there is no significant change in β.

The susceptibility exponent γ is dramatically larger in
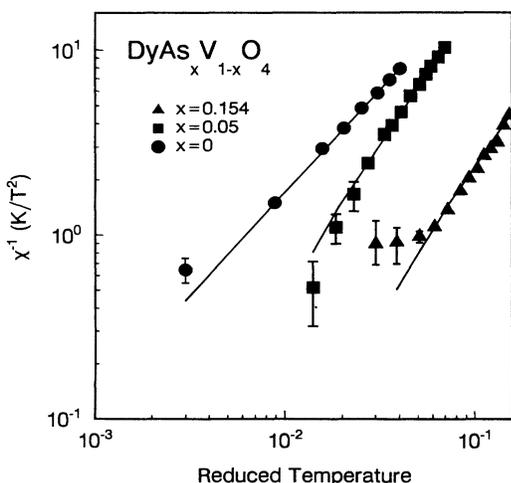
FIG. 8. Log-log plots of inverse susceptibility vs reduced temperature for the three samples. Table I gives values for γ obtained from the slopes.

the mixed compounds. However, our result for the $x = 0.154$ sample, $\gamma = 1.57 \pm 0.06$, is somewhat less than the value 1.79 ± 0.07 obtained in ultrasonics measurements.[6] One should be cautious about concluding that there is a discrepancy between these two measurements in view of the different techniques used. The present experiments essentially provide a static measurement of γ, while the ultrasonic experiments use frequencies of 10–100 MHz. Frequency dependence due to critical rounding was looked for in the ultrasonics experiments,[20] but it appeared to be a small effect. However either result may be affected by other systematic errors: it is difficult, in general, to ensure their absence in the measurement of critical exponents. At any rate, our value for γ is a reasonable one in the context of theoretical and numerical estimates of γ for the RFI model, which, although somewhat scattered, lie in the range from 1.5 to 2.0.[1]

We observe a significant increase in δ in the mixed compounds, although our measurements are not very precise as mentioned above. Since T_D is not known accurately during the experiments, one is unlikely to get a set of data at exactly T_D. However, we analyzed data obtained at temperatures slightly above and below T_D and found that they gave very similar results for δ.

The failure of β to change in the mixed samples is surprising, but is confirmed by measurements on different mixed samples and by the use of two different techniques, one in the twinned phase and one in the single-domain phase. Moreover, our values for β in pure DyVO$_4$ agreed with the expected result, and an additional experiment on pure TbVO$_4$ (see Fig. 5) gave $\beta = 0.47$, close to the expected value of 0.5.[12]

While there is no accepted prediction of β for the RFI model, both theoretical and numerical studies[1] have predicted that β should be very small or even zero (first-order transition). There is some experimental support, although not conclusive, for a reduced value for β from experiments on DAF systems. Thurston et al.[21] measured $\beta = 0.21 \pm 0.07$ in a system where the random fields were weak and hysteresis was not observed; however, given the weakness of the random fields they suggested that their value represented an average of RFI and REI (random-exchange Ising) exponents. Ramos et al.[10] reported results in rather stronger random fields, but because of equilibration problems could only give an upper limit, $\beta \gtrsim 1/8$. Very recently Sartorelli[22] analyzed fluorine NMR line shapes in Fe$_{0.46}$Zn$_{0.54}$F$_2$ to extract a value for $\beta = 0.125$ in the random-field regime. It is difficult to assess the accuracy of this result, however, since the analysis depends on determining the second moment of the resonance line in a narrow temperature range where the line shape changes from Gaussian to Lorentzian and back to Gaussian. On the other hand, a recent measurement[23] of β for the liquid-vapor transition in a porous medium, another system that is believed to a realization of the RFI model, obtained the result $\beta = 0.28 \pm 0.05$. It was subsequently suggested[24] that the presence of asymmetric random fields in this system implies that the universality class is different from that for the simple RFI model.

Our ability to measure the three exponents β, δ, and γ in the same sample allows us to test whether the scal-

ing law of Eq. (1) is satisfied for both pure and mixed $Dy(As_xV_{1-x})O_4$ samples as must be the case if they belong to distinct universality classes. Since knowledge of any two exponents permits calculation of the third, we show a comparison in Table II between δ calculated from $1 + \gamma/\beta$ and the measured values of δ. The two values of δ are seen to agree well for both pure $DyVO_4$ and the mixed crystals, in support of the conclusion that these systems belong to distinct universality classes.

As yet we have not measured the specific-heat exponent α and therefore cannot test the well-known scaling relation $\alpha + 2\beta + \gamma = 2$. On the other hand our results for β, δ, and γ seem consistent and allow us to calculate and compare α for the pure and mixed systems. The calculated values are shown in Table II and predict a significant change in the critical specific heat from the pure (divergent, $\alpha \simeq 0.2$) to the mixed (cusp, $\alpha \simeq -0.2$) samples. It is worth noting that this behavior is consistent with the Harris criterion[25] for the conditions under which the pure and disordered systems have sharp phase transitions in distinct universality classes.

Although our results appear to be reliable and self-consistent, there are major concerns about their interpretation in the context of what is known or believed about the RFI model. The apparent discrepancy between our value for β and the expected much smaller value is one problem. Another is the inference of a specific heat cusp rather than the logarithmic divergence ($\alpha = 0$) observed in DAF experiments.[2] It is certainly reasonable, therefore, to reexamine the basis for identifying this system as a RFI model. We have argued that the As-V substitutions do not affect the Dy-Dy interactions, and that they therefore introduce random fields and not random exchange. These interactions, however, are mediated by distortions[5] of the lattice of atoms and any change in atomic mass will modify the interactions to some extent. Thus, in principle, there could be some component of REI model behavior in the observed critical properties in our

TABLE II.   Critical exponents δ and α for $Dy(As_xV_{1-x})O_4$ calculated from scaling relations.

x	δ (calc.)	δ (meas.)	α (calc.)
0	4.4(4)	4.1(5)	0.18(11)
0.050	5.9(5)	5.5(5)	-0.14(12)
0.154	5.8(6)	6.1(7)	-0.22(13)

system. On the other hand, it is difficult to understand the sharply reduced transition temperatures (by $\sim 40\%$ for $x = 0.154$) or the complete suppression of the transition for larger values of x, in terms of random exchange rather than random-field effects. Although our values for β are quite compatible with the REI model, our values for γ are well above the expected REI model result (see Table 1 of Ref. 2). Another point has been raised by Toh[26] who argued that the anisotropic mode softening in structural phase transitions with random strains leads to different critical behavior from that of the simple RFI model. Our results, however, are not in accord with the critical exponents he has calculated.

In summary, we have found that the critical exponents β, δ, and γ for a structural Ising-model phase transition are altered by the presence of random strains. The new exponents satisfy the scaling relation $\gamma = \beta(\delta - 1)$ but are not consistent with expectations of the exponents for the RFI model. The most striking result of our measurements is that there is no sign of a decrease in β for quite strong random fields. Future experiments will attempt to confirm and extend these measurements and to investigate effects of random strains in other structural phase transitions.

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