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ULTRASONIC INVESTIGATION OF MODIFIED CRITICAL BEHAVIOUR IN THE STRONG RANDOM-FIELD SYSTEM Dy (As_{0.17}V_{0.83}) O₄

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Abstract. - Ultrasonic measurements of the soft-mode elastic constant $1/2(C_{11} - C_{12})$ have been performed to investigate the pseudospin susceptibility of the random-field system Dy (As_xV_{1-x}) O₄. The susceptibility critical exponent $\gamma = 1.73 \pm 0.07$ for $x = 0.17$ is much larger than for the pure compound ($x = 0$), in reasonable agreement with predictions of the random-field Ising model.

While much of the recent interest in the effects of random fields on Ising phase transitions has focussed on magnetic systems, some of the most important features of this problem - the new critical behaviour and the slow equilibration near and below the transition temperature - can also be studied experimentally in mixed Jahn-Teller (JT) compounds [1]. The structural phase transitions in these materials [2] are driven by a linear coupling between a lattice distortion and electronic energy levels of rare-earth or transition ions, giving rise in most cases to a ferrodistorptive effective ion-ion interaction that is very well described by a pseudospin Ising Hamiltonian $-\sum J_{ij} \sigma_i^z \sigma_j^z$. In mixed crystals, the ionic size mismatch generates static random strain fields, one component of which has the right symmetry to couple to the electronic operators σ_i^z and give the additional random-field (RF) term $-\sum h_i \sigma_i^z$ in the pseudospin Hamiltonian. Thus mixed JT compounds are the structural counterparts of a ferromagnet in a random magnetic field, and are potentially important since this magnetic system cannot be realized in practice, all magnetic experiments having been performed on diluted antiferromagnets in a uniform field.

Of the mixed JT systems studied so far, Dy (As_xV_{1-x}) O₄ is probably the most interesting in the context of RF effects: the JT interactions in the parent compounds DyVO₄ and DyAsO₄ are of comparable magnitude [3], the interactions are not diluted on mixing so that only the RFs should depend on concentration x , and the interactions have a large short-range component leading to non-classical critical behaviour in the range of temperatures that are normally accessible experimentally [3, 4]. Indeed our previous electric susceptibility and ultrasonic measurements [1] in these compounds have shown evidence of metastable ordering, history dependence and modified critical behaviour that includes some previously unexplored features of random-field Ising systems. In this paper we present the results of new ultrasonic velocity measurements that were performed to obtain better data on

the critical behaviour of both mixed ($x = 0.17$) and pure ($x = 0$) samples of Dy (As_xV_{1-x}) O₄.

In these Dy compounds, the lattice distortion is from tetragonal to orthorhombic symmetry corresponding to the soft acoustic shear mode with elastic constant $1/2(C_{11} - C_{12})$. Hence just above the distortion temperature T_D , the softening of $1/2(C_{11} - C_{12})$ is inversely proportional to the divergence of the pseudospin susceptibility, i.e. $1/2(C_{11} - C_{12}) \propto \chi^{-1} \propto t^\gamma$ where $t = (T - T_D)/T_D$ is the reduced temperature and γ is the susceptibility critical exponent. As in our earlier work [5], we have determined γ from the ultrasonic velocity of [100] longitudinal waves by subtracting the temperature-independent contribution of $1/2(C_{11} + C_{12})$ from the measured elastic constant $\rho v^2 = C_{11} = 1/2(C_{11} + C_{12}) + 1/2(C_{11} - C_{12})$. (This trick avoids the prohibitively large ultrasonic attenuation encountered near T_D when $1/2(C_{11} - C_{12})$ is measured directly using [110] shear waves.) Figure 1 shows the temperature dependence of C_{11} for the mixed and pure samples from 4 to 80 K. As the temperature is lowered, C_{11} decreases continuously until a mini-

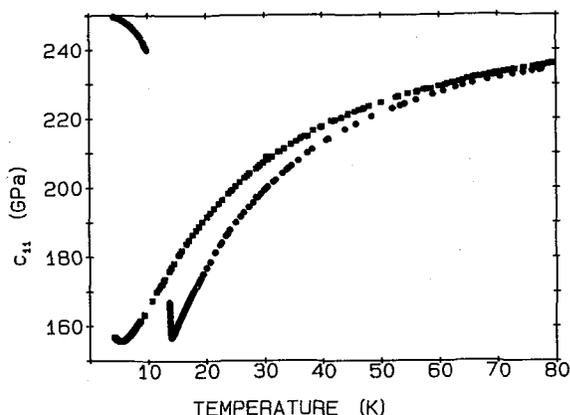


Fig. 1. - Elastic constant C_{11} vs. temperature for Dy (As_xV_{1-x}) O₄ with $x = 0.17$ (■) and $x = 0$ (●).

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imum is reached at the phase transition temperature for each sample, implying a second-order phase transition in both cases. From the values of T_D (5.57 K and 14.06 K for the mixed and pure samples respectively) it follows that the transition temperature in the mixed crystal is 40 % that of pure DyVO_4 , indicating that the random fields are strong. The figure also shows that the random fields cause the minimum of C_{11} to be less sharp but more symmetric in the mixed material. Within experimental error, both the degree of softening of C_{11} and the values of C_{11} at T_D are the same for both samples, consistent with the result that $1/2(C_{11} - C_{12}) = 0$ at the phase transition for $\text{Dy}(\text{As}_{0.17}\text{V}_{0.83})\text{O}_4$ as well as for pure DyVO_4 . Thus we have taken $1/2(C_{11} + C_{12}) = C_{11}(T_D)$ in determining the temperature dependence of $1/2(C_{11} - C_{12})$ from our data. This is further justified by the good agreement between these values of $C_{11}(T_D)$ and $1/2(C_{11} + C_{12})$ measured in another rare earth zircon TmVO_4 [6].

Figure 2 shows a log-log plot of $1/2(C_{11} - C_{12})$ versus reduced temperature t for the two samples. In pure DyVO_4 , reasonable power-law behaviour is found for $10^{-2} < t < 10^{-1}$ with slope $\gamma = 1.13 \pm 0.07$. This value confirms that mean field behaviour ($\gamma = 1.0$) is not observed in this range of reduced temperatures, but it is somewhat smaller than the Ising exponent $\gamma = 1.25$ expected from earlier birefringence data [4]. The deviations from this power law seen at lower t , where possible errors in T_D and in the zero of the elastic constant have greatest effect, may simply re-

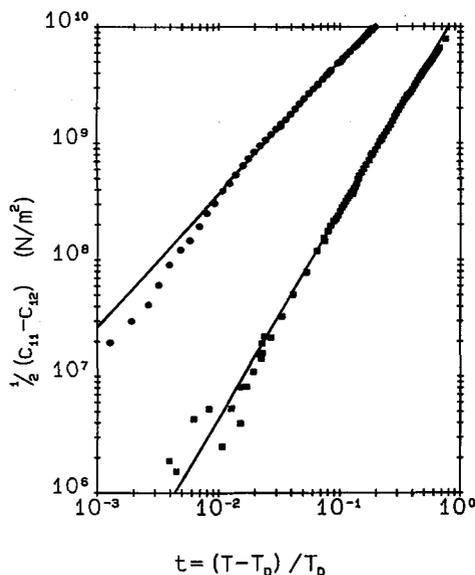


Fig. 2. - Elastic constant $1/2(C_{11} - C_{12})$ vs. reduced temperature for $\text{Dy}(\text{As}_x\text{V}_{1-x})\text{O}_4$ with $x = 0.17$ (\blacksquare) and $x = 0$ (\bullet). The straight lines have slopes 1.73 and 1.13 as discussed in the text.

sult from weak random fields due to a small amount of flux impurities; we note that T_D in this sample is about 4 % lower than in our best samples studied by electric susceptibility measurements [3]. Nonetheless it is clear from figure 2 that γ appears to be much larger in the mixed crystal $\text{Dy}(\text{As}_{0.17}\text{V}_{0.83})\text{O}_4$, with $\gamma = 1.73 \pm 0.07$ fitting the data over nearly two orders of magnitude in reduced temperature. This RF exponent $\gamma = 1.73$ is consistent with the value 1.75 expected if RF critical behaviour is correctly described by a dimensionality reduction of 1 [7], with the value 1.75 ± 0.02 observed in experiments on dilute antiferromagnets [8], and with the value 2.0 ± 0.5 found in the most recent simulations [9]. However, the exponent $\gamma = 1.73$ apparently disagrees with recent scaling predictions for the RF Ising model in $2 + \epsilon$ dimensions [10] which give $\gamma = (3 - d + 1.5\epsilon) / \epsilon = 1.5$ for $d = 3$, $\epsilon = 1$. A detailed comparison with this and other work may nevertheless be complicated by possible correlations between the random fields at different sites and the effects of transverse random fields of either A_{1g} or B_{2g} symmetry. Future experimental work will further investigate possible effects of concentration gradients in the mixed sample ($\Delta x/x < 10^{-3}$ over 2.5 mm in typical samples [1]) and combine shear wave measurements of $1/2(C_{11} - C_{12})$ with the longitudinal ultrasonic data to obtain a more definitive check on the correct zero of the elastic constant.

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