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Static and dynamic critical effects in the random-field system $Dy(As_xV_{1-x})O_4$

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Dynamic rounding of the susceptibility in strong Random-Field (RF) samples of $Dy(As_xV_{1-x})O_4$ has been observed in the critical region using ultrasonic velocity measurements. In weaker random fields, however, the dynamics do not mask the static critical behaviour, enabling an accurate measurement, $\gamma = 1.80 \pm 0.07$, to be made of the RF susceptibility critical exponent.

Over the last few years, ultrasonic [1], neutron scattering [2], electric susceptibility [3] and optical [4] measurements have shown that the mixed Jahn–Teller compounds $Dy(As_xV_{1-x})O_4$ are an interesting experimental system in which to study the unusual critical behaviour and slow equilibration properties of the Random-Field Ising Model (RFIM). These mixed compounds undergo structural phase transitions that are driven by ferrodistortive Ising interactions in the presence of random strain fields generated by the As/V size mismatch; they are therefore structural counterparts of an Ising Ferromagnet in a random magnetic field. We have previously shown [1] that ultrasonic velocity measurements can be used to obtain a precise value of the susceptibility exponent γ and that this exponent is significantly increased by Random Fields (RFs), giving clear evidence that the RFIM belongs to a new universality class. Here we present the results of new ultrasonic experiments to look for dynamic critical effects caused by the random fields and to investigate in more detail the background contribution to the elastic constant from which the susceptibility is determined.

Near the transition temperature, the susceptibility of $Dy(As_xV_{1-x})O_4$ is inversely proportional to the soft-mode elastic constant $\frac{1}{2}(c_{11} - c_{12})$ which we have measured using longitudinal [100] ultrasonic waves ($\rho v^2 = c_{11} = \frac{1}{2}(c_{11} + c_{12}) + \frac{1}{2}(c_{11} - c_{12})$, where ρ is the density and v the velocity), the ultrasonic attenuation being too high to investigate the soft-mode critical behaviour directly using shear waves. Since $\frac{1}{2}(c_{11} + c_{12})$ remains almost constant at low temperatures (see below), the behaviour of c_{11} is dominated by the variation of $\frac{1}{2}(c_{11} - c_{12})$, thus enabling the critical behaviour of the susceptibility to be investigated. Figure 1 shows the temperature dependence of c_{11} at ultrasonic frequencies between 25 and 120 MHz in three samples of $Dy(As_xV_{1-x})O_4$ with different random-field strengths. It can be seen that in sample A no frequency dependence of the elastic constant is observed [5], whereas in samples B1 and B2 the minimum in c_{11} becomes shallower and broader as the frequency is increased, indicating that dynamic effects due to slowing down of the critical fluctuations as T_D is approached are relatively important in the ultrasonic frequency range only for these stronger RF samples. These results demonstrate clearly that the critical slowing down is greatly enhanced by random fields, as has been found experimentally in RF magnets [6] and predicted theoretically for the RFIM by the activated dynamics models of Villain [7] and Fisher [8]. In these models the characteristic time τ for a fluctuation on a scale of the correlation length ξ grows exponentially as $\tau \alpha \exp(\xi^{\theta})$, with the result that the dynamic scaling of the elastic constant can be written $c(\omega,t) \alpha t^{\gamma} \cdot F\{|\ln(\omega/\omega_0)|/t^{-\theta\nu}\}$, where t is the reduced temperature, F is a universal scaling function, ω_0 is some characteristic frequency, θ is a new exponent that governs the growth of the free energy in a volume ξ^d and ν is the correlation length exponent. While this expression for $c(\omega,t)$

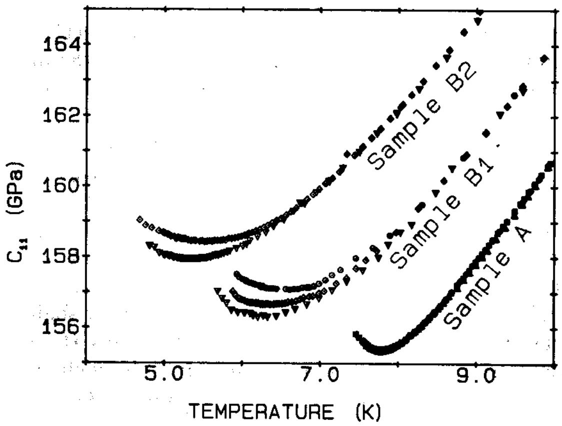

Fig. 1. Temperature dependence of c_{11} in three samples of $Dy(As_xV_{1-x})O_4$ at selected ultrasonic frequencies: 25 MHz (▼), 30 MHz (▲), 50 MHz (■), 68 MHz (◇), 70 MHz (●) and 120 MHz (○). The arsenic concentrations x and transition temperature reduction ratios $r = T_D/T_D$(pure) for these samples are: $x = 0.154 \pm 0.01$ and $r = 0.54$ for sample A, $x = 0.164 \pm 0.01$ and $r = 0.42$ for sample B1, and $x = 0.164 \pm 0.01$ and $r = 0.38$ for sample B2. (Note that the difference in r between samples B1 and B2 implies a slight difference also in x, but this was too small for us to measure.)

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gives a good description of our data using reasonable values of the scaling-law parameters (e.g. $\nu\theta = 1.05$ as in ref. [6], $\gamma = 1.79$ from our work [1], $200 \leq \omega_0/2\pi \leq 1000$ MHz), data spanning a much wider range of frequencies are needed to determine whether an activated model gives a better description of the dynamics in $Dy(As_xV_{1-x})O_4$ than conventional dynamic scaling (cf. Ref. [6]) and to determine the scaling-law parameters precisely.

For sample A, in which the soft-mode elastic constant was found to be independent of frequency between 10 and 70 MHz, the static critical behaviour is not obscured by the dynamic effects seen in the other samples, enabling the susceptibility exponent γ to be measured from the relation $\frac{1}{2}(c_{11} - c_{12}) \alpha \chi^{-1} \alpha t^{\gamma}$. To determine the soft-mode elastic constant from the data shown in fig. 1, we subtracted from c_{11} the background contribution $\frac{1}{2}(c_{11} + c_{12})$, obtained from the difference between ρv^2 for longitudinal [110] waves (this gives $c_L = \frac{1}{2}(c_{11} + c_{12}) + c_{66}$) and for shear [100] waves (this gives c_{66} only). The variations of c_L and c_{66} with temperature are shown in fig. 2. Although the c_{66} is virtually independent of temperature up to 20 K, indicating that the coupling to the B_{2g} mode is extremely weak, the elastic constant $\frac{1}{2}(c_{11} + c_{12})$ softens slightly near T_D, implying that the coupling to this (totally symmetric) A_{1g} strain mode is not entirely negligible. Physically this A_{1g} coupling induces a small dilation or contraction of the tetragonal unit cell in the basal plane, and while the possibility of a measurable A_{1g} coupling in these Dy compounds has been recognized for some time, this is the first time that it has been observed experimentally. The soft-mode elastic constant $\frac{1}{2}(c_{11}\text{-}c_{12})$ obtained from the data of figs. 1 and 2 is plotted versus reduced temperature in the log–log graph of fig. 3, where we have used the fact that $\frac{1}{2}(c_{11} - c_{12})$ must go to zero at T_D in the static limit to establish the correct baseline, the absolute uncertainty

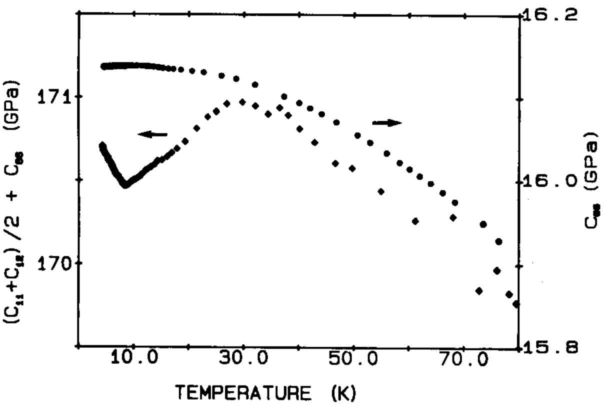

Fig. 2. Temperature dependence of the measured elastic constants from which $\frac{1}{2}(c_{11} + c_{12})$ was determined for sample A. The data $\frac{1}{2}(c_{11} + c_{12}) + c_{66}$ have been corrected for a small misalignment of the [110] axis of $1.2 \pm 0.7°$, introducing an additional uncertainty of less than ± 0.05 GPa in the elastic constant.

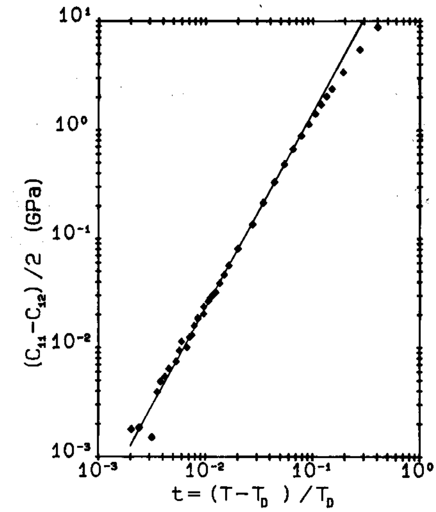

Fig. 3. $\frac{1}{2}(c_{11} - c_{12})$ vs reduced temperature t for sample A.

in the difference between c_{11} and $\frac{1}{2}(c_{11} + c_{12})$ being too large (± 1.2 GPa) to determine this directly from our measurements. From the slope of this graph we obtain $\gamma = 1.80 \pm 0.07$, which agrees exactly with our earlier value for this sample obtained without correcting for the weak temperature dependence of $\frac{1}{2}(c_{11} + c_{12})$. It is interesting that our result for the RF susceptibility exponent in $Dy(As_xV_{1-x})O_4$ is consistent with the two-dimensional nearest-neighbour Ising value $\gamma = 7/4$ and with simulations of the short-range RFIM, but is in disagreement with certain scaling predictions. A more complete discussion of these points is given in ref. [1], where it is also shown that the effects of concentration inhomogeneities on our measurement of γ are negligible.

In conclusion we note that neither of the new results presented in this paper (viz. the observation of dynamic critical effects and the discovery of a weak anomaly in $\frac{1}{2}(c_{11} + c_{12})$ near T_D) affects our earlier result for γ, confirming that our ultrasonic measurements in $Dy(As_xV_{1-x})O_4$ have yielded the most reliable value thus far of this RFIM exponent.

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