Absence of true critical exponents in relaxor ferroelectrics: the case for nanodomain freezing

2006 J. Phys.: Condens. Matter 18 L523

(http://iopscience.iop.org/0953-8984/18/41/L03)

J. Phys.: Condens. Matter **18** (2006) L523–L526 doi:10.1088/0953-8984/18/41/L03

LETTER TO THE EDITOR

Absence of true critical exponents in relaxor ferroelectrics: the case for nanodomain freezing

Wolfgang Kleemann

Angewandte Physik, Universität Duisburg-Essen, 47048 Duisburg, Germany

Received 28 August 2006
Published 29 September 2006
Online at stacks.iop.org/JPhysCM/18/L523

Abstract

Scott's (2006 *J. Phys.: Condens. Matter* **18** 7123) recent review of Kleemann *et al*'s (2002 *Europhys. Lett.* **57** 14) critical exponents of strontium barium niobate is shown to be misled by erroneous input parameters. Although the observed set of exponents reflects, indeed, the absence of true three-dimensional (3D) random-field Ising model critical behaviour, it cannot be compatible with the proposed domain wall model in $d = 2.5$ dimensions or with Levanyuk and Sigov's (1988 *Defects and Structural Phase Transitions* (London: Gordon and Breach)) defect model. As was argued independently by Kleemann *et al* (2006 *Phys. Rev. Lett.* **97** 065702), it is rather in agreement with the pure two-dimensional (2D) Ising model.

The claimed discovery of the first materialization of the ferroic three-dimensional (3D) random-field Ising model (RFIM) system by the uniaxial relaxor ferroelectric strontium barium niobate (SBN61, $Sr_{0.61}Ba_{0.39}Nb_2O_6$, and its Ce^{3+}-doped descendants) [1] left open the question of how to understand the critical exponents observed. Hence, not unexpectedly, a controversial discussion about their very relevance immediately started and recently culminated in a very involved discussion by Scott [2], who (i) stated that the exponents observed [1] cannot reflect true equilibrium critical behaviour, and (ii) simultaneously proposed two alternative interpretations. He proposed either possible critical behaviour in $d = 2.5$ dimensions on domain walls or an agreement with Levanyuk and Sigov's [3] defect model.

In this letter we confirm on the one hand that the first statement is, indeed, correct. On the other hand, however, we show that the above discussion was partially misled by erroneous citations and that the models proposed are not compatible with the experimental observations. Our arguments are based on very new dielectric and scanning probe results [4], which suggest that SBN freezes into nanopolar regions at temperatures close to $T = T_c$. Consequently, we rather propose a pure Ising model in $d = 2$ dimensions to account for the criticality remaining active within the interfaces between the polar nanoregions [4].

Table 1 shows a list of the critical exponents α, β and γ measured for SBN61 using different methods, as indicated. They are compared with those obtained for the diluted uniaxial antiferromagnet $Fe_{1x}Zn_xF_2$ in a homogeneous axial magnetic field (DAFF), which is known to

Table 1. Static critical exponents measured for the relaxor SBN61 and the DAFF system $Fe_{1-x}Zn_xF_2$ in comparison with theoretical values for the 3D RFIM and the 2D Ising model.

	Experiment		Theory	
Critical exponent	Relaxor SBN61	DAFF $Fe_{1-x}Zn_xF_2$	3D RFIM	2D Ising
Specific heat, α	-0.02 ± 0.02 [4]	≈ 0 [6]	-0.63 ± 0.07 [9]	0 [14]
Order parameter β	0.13 ± 0.02 SHG [4] 0.14 ± 0.03 NMR [5]	0.16 ± 0.02 [7]	0.017 ± 0.005 [10] 0.02 ± 0.01 [11]	0.125 [15]
Order parameter susceptibility, γ	1.85 ± 0.15 [1]	1.58 ± 0.13 [8]	1.7 ± 0.2 [12] 1.5 ± 0.2 [13]	1.75 [14]

belong to the same universality class as the RFIM [16]. While the exponents of experimentally realized RFIM systems (SBN and SBN:Ce^{3+}) come close to those obtained on the prototypical DAFF system $Fe_{1-x}Zn_xF_2$, a comparison with theoretical values for the 3D RFIM in table 1 reveals significant deviations. It has to be noticed that the latter values are still a matter of debate and only those from the most recent sources are listed. Beyond errors, however, serious discrepancies arise in particular for the exponents of the specific heat, α, and of the order parameter, β. The values $\alpha \approx 0$ and $\beta \approx 0.13$–0.16 extracted from the experiments contrast sensitively with $\alpha \approx -0.6$ and $\beta \approx 0$, according to theory and simulations. Much better agreement is stated when comparing the experimental results with those of the unperturbed ('pure') 2D Ising model, whose exact critical exponents have been well known for a long time (table 1). Comparison of the experimental values of the exponent γ between experiment and theory (values depending significantly on the random field distribution!) does not allow for an unambiguous distinction between 3D RFIM and 2D Ising.

This surprising coincidence has been discussed for the DAFF exponents in the 1980s and the seeming 'reduction of dimensionality' became a matter of heavy debate among theoreticians [17]. One major discovery was the so-called violation of hyperscaling, which explicitly connects the critical exponents with the spatial dimension d. A new scaling exponent $\theta \approx 1.5$ was introduced for the temperature at the $T = 0$ fixed point [18, 19], which is now generally accepted. However, this rigorous result could also not solve the above discrepancies. In other words, a 'projection' from the '3D random-field' to the '2D pure' Ising model has never been justified on any serious theoretical ground.

Only very recently we proposed that the long-standing puzzle can be solved when considering the observed self-organized subdivision of the ferroelectric RFIM systems SBN and SBN:Ce^{3+} into metastable ('frozen') domains embedded in a network of quasi-2D interfaces [4]. While the 'domains' experience polar long-range order already above the phase transition temperature, T_c, by virtue of the spatial fluctuations of quenched random fields, the 'interfaces' are subject to quasi-staggered fields, which do not give rise to ferroic correlations on a mesoscale. A similar subdivision, albeit only for temperatures $T < T_c$, was considered by Middleton and Fisher [10] in order to interpret some of the results of their simulations for the RFIM. Quite naturally, we proposed that the apparent 2D Ising criticality actually takes place on the 2D subspace of the interfaces. It is thus a consequence of the giant critical slowing-down of the RFIM [19] which finally hampers the observability of true 3D RFIM critical exponents when approaching T_c on 'normal' laboratory timescales. For example, we have proven the stability of frozen bulk domains at reduced temperatures as large as $T/T_c - 1 \approx 0.02$ on a timescale of $\tau \approx 10^3$ s by scanning piezoforce microscopy (PFM) [4].

Virtually simultaneously with our publication [4], submitted on 12 May 2006, alternative interpretations of our experimental critical exponents were proposed in a paper by Scott [2], submitted on 16 May 2006. Basically, he also comes to the conclusion that true critical behaviour might be unobservable in the RFIM owing to its tendency to become stuck in metastable configurations. In order to 'understand' the observed exponents within the framework of an extensive scaling analysis, he first proposes another effective reduction of symmetry on the basis of a domain wall model with a fractal dimension $d = 2.5$ without justifying or explaining, however, the origin of domain walls at $T > T_c$. His second proposition refers to a defect model due to Levanyuk and Sigov [3]. It is easily seen that both of these propositions fail in reproducing our critical exponents. In particular, the unanimously reported value $\alpha \approx 0$ [4, 6] disagrees sharply with all the values, $\alpha > 0$, quoted in table 1 of [2]. Also, the value $\beta \approx 1/8$, as essentially confirmed by all of the pertinent experiments [4, 5, 7], lies far below the range offered by Scott's models [2], $1/4 \leqslant \beta \leqslant 1/2$. We note that the low value $\beta = 0.06$ argued by Scott [2] to be one of our early experimental results was in reality another theoretical result [20][1].

The Levanyuk–Sigov model [3] was cautiously favoured [2], since its low value of the critical isotherm exponent, $\delta = 2$, seems to come close to an exponent $\delta = 1.53 \pm 0.15$ published by ourselves [21]. However, this comparison is a matter of a serious misunderstanding. The critical isotherm exponent, δ, which Scott has in mind was never measured in SBN because of extreme equilibration problems arising at T_c [23]. In reality, the above-cited exponent is the size distribution exponent of polar domains in the ferroelectric state of SBN [21]. Unfortunately, it bears the same name, where $\delta = 1.53$ enters the formula $N(A) \propto A^{-\delta}$, N being the number of domains and A the domain cross section [21]. Another misunderstood number used in Scott's discussion [2] is the 'fractal dimension of the ferroelectric domain walls' in SBN. On the occasion of IMF-11 (Iguazu, Brazil, August 2005), I introduced a scaling ansatz for the density of states of pinned domain wall segments with length L, $g(L) \propto L^{-x}$. A relationship of the exponent x to the fractality of the domain wall was mentioned and published [22], where the emerging values, $1 \leqslant x \leqslant 1.7$, might be taken as a 'fractal dimension' of the domain wall contour line. This 'fractal dimension', referring to the projection of the wall onto the plane of observation, must, however, not be confused with the fractal dimension of the domain wall area, which is undoubtedly $d^* \geqslant 2$.

In conclusion, Scott's [2] discussion of the hitherto poorly understood experimental critical exponents of the first experimental realization of the ferroic 3D RFIM, the relaxor ferroelectric SBN [1], independently confirms our result [4], *viz.* that asymptotic criticality is unobservable due to severe thermal equilibration problems. His inferences, however, with respect to a possible alternative physical model for the metastable system are not convincing and suffer from flaws based on input data taken incorrectly from the literature. Instead, we propose another more plausible solution of the RFIM criticality enigma on the grounds of recent structural investigations, which suggest that the criticality of a metastable 2D Ising model is involved [4].

References

[1] Kleemann W, Dec J, Lehnen P, Blinc R, Zalar B and Pankrath R 2002 *Europhys. Lett.* **57** 14
[2] Scott J F 2006 *J. Phys.: Condens. Matter* **18** 7123
[3] Levanyuk A P and Sigov A S 1988 *Defects and Structural Phase Transitions* (London: Gordon and Breach)
[4] Kleemann W, Dec J, Shvartsman V V, Kutnjak Z and Braun T 2006 *Phys. Rev. Lett.* **97** 065702
[5] Blinc R, Gregorovic A, Zalar B, Pirc R, Seliger J, Kleemann W, Lushnikov S G and Pankrath R 2001 *Phys. Rev.* B **64** 134109

[1] Their value $\beta = 0.06$ for the RFIM order parameter was quoted by the author on the occasion of ICDMRS (Hernosissos, Crete, June 2001), as published by Kleemann [20].

[6] Slanic Z and Belanger D P 1998 *J. Magn. Magn. Mater.* **186** 65
[7] Ye F, Zhou L, Larochelle S, Lu L, Belanger D P, Greven M and Lederman D 2002 *Phys. Rev. Lett.* **89** 157202
[8] Slanic Z, Belanger D P and Fernandez-Baca J-A 1999 *Phys. Rev. Lett.* **82** 426
[9] Hartmann A K and Young A P 2001 *Phys. Rev.* B **64** 214419
[10] Middleton A A and Fisher D S 2002 *Phys. Rev.* B **65** 134411
[11] Hartmann A K and Nowak U 1999 *Eur. Phys. J.* B **7** 105
[12] Rieger H 1995 *Phys. Rev.* B **52** 6659
[13] Nowak U, Usadel K D and Esser J 1998 *Physica* A **250** 1
[14] Onsager L 1944 *Phys. Rev.* **65** 117
[15] Yang C N 1952 *Phys. Rev.* **85** 808
[16] Fishman S and Aharony A 1978 *J. Phys. C: Solid State Phys.* **12** L729
[17] Belanger D P and Young A P 1991 *J. Magn. Magn. Mater.* **100** 272
[18] Bray A J and Moore M A 1985 *J. Phys. C: Solid State Phys.* **18** L927
Villain J 1984 *J. Physique* **46** 1843
[19] Fisher D S 1985 *Phys. Rev. Lett.* **56** 416
[20] Newman M E J and Barkema G T 1996 *Phys. Rev.* E **53** 393
Kleemann W 2002 *J. Non-Cryst. Solids* **307** 66
[21] Lehnen P, Kleemann W, Woike T and Pankrath R 2001 *Phys. Rev.* B **64** 224109
[22] Kleemann W, Dec J, Prosandeev S A, Braun T and Thomas P A 2006 *Ferroelectrics* **334** 3
[23] Granzow T 2005 private communication