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Analysis of TbZn resistivity temperature derivative above the Curie point using singular fitting equations

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Abstract

New analyses were carried out of previously published TbZn resistivity temperature-derivative data just above the Curie temperature, T_c . Fitting was accomplished using a weighted nonlinear least-squares program, GENLS, which allows one to take proper account of errors present in both the independent and the dependent variables. The utility of fitting with equations of either logarithmic or power-law form (involving a critical exponent λ), both with a pole at the Curie point, was investigated in detail. Fitting results were uncertain for the four-parameter power-law choice but, with one parameter fixed, led to estimates of T_c of 199.61 ± 0.02 K and of λ of 0.16 ± 0.01 . The latter value is rather far removed from the value of 0.104 which is expected for a three-dimensional ($n = 1$) Ising model and from the value of 0.014 for a three-dimensional ($n = 2$) XY model. By contrast, fitting with a three-parameter logarithmic law, corresponding to a three-dimensional ($n = 2$) XY model, led to estimated standard deviations of all three free parameters much smaller than the corresponding ones of the four-parameter power-law fit and to a T_c estimate of 199.56 ± 0.03 K. The present results thus indicate the presence of marked XY or even Ising anisotropy. Quadrupolar effects seem irrelevant.

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Key words: Critical exponent; Electrical resistivity; TbZn; Curve fitting; Magnetic model

1. Introduction and background

A number of studies have been published on critical-point singularities in fluids and in magnets. Historically, the works of Ahlers on the specific heat singularity at the superfluid transition [1] and those of Vicentini-Missoni et al. [2] on the β and δ critical exponents and the equation of state [3] were crucial. The situation is more

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complicated when the physical property remains finite on both sides of the transition temperature. This is the case of the electrical resistivity in metallic magnets, for example [4]. A “background term” has to be taken into account, with the complication that the “background” contains at least some mean field contribution from the order parameter itself below the critical temperature. Therefore, it is difficult to disentangle the truly critical singular term, one is looking for from the measured behavior. The best resort is to take the derivative of the physical property up to the order at which one obtains a divergence, the so-called λ -like transition. The first derivative is enough for metallic magnets [4].

Then, an analysis of the resistivity(ρ)–temperature (T) data near a critical point can, in principle, yield valuable information about the origin of the phase transition. However, the problem of obtaining good estimates of the parameters of a fitting model, such as the critical temperature and the critical exponent, is not yet entirely straightforward because of the presence of the critical-point singularity itself [5]. One of the best-test cases to our knowledge lies in the excellent resistivity–temperature data for the magnetic material TbZn obtained and analyzed by Sousa et al. [6]. Some very careful painstaking work allowed them to use very closely spaced temperature values in order to numerically differentiate ρ values and obtain reliable data for the resistivity derivative, $d\rho/dT = \rho'$. Particular attention was devoted to the temperature region just above the Curie point, T_c .

By fitting such (ρ' , T) data to various equations which have a singularity at $T = T_c$, one can attempt to estimate such important properties of the phase transition as its critical exponent, λ , and T_c itself. The above data were later re-analyzed [4,7] with different fitting equations and methods. But in none of these fittings were estimates of parameter standard deviations presented, making the choice of the most appropriate fitting equation uncertain, as well as the validity of the parameters themselves. Therefore, no definite physical conclusion could be reached concerning the order-parameter symmetry at the transition.

Fortunately, more definite conclusions are possible using a different and more powerful fitting procedure than those employed earlier. Suppose that one has carried out a new fit of precise, high-resolution data to an appropriate singular-response model (see below) one will obtain not only an estimate of T_c and of the leading singularity but also an estimate of the standard deviation, s_{T_c} . Only after this, will it then be natural to ask whether scaling corrections to the fitting model are needed when individual temperature measurements fall inside or near the range ($T_c - s_{T_c}$) to ($T_c + s_{T_c}$), where these limits involve the estimated values. Notice that such corrections have indeed been calculated previously for anisotropic (anti- and ferro-) magnetic metals by one of us [8], following an earlier analysis of the spin–spin correlation function [9]. It was found that the leading correction term involved a positive critical exponent θ , estimated to be 0.5 [7], which would lead to a vanishing correction at the critical temperature itself.

There is, however, general agreement that the possibility of isolating a vanishingly small power-law term from a finite dominant term is questionable. Therefore, we

avoid such an investigation of θ here, concentrating on obtaining the first (most singular) term in ρ' . For these reasons, we have omitted any scaling corrections in the present work. Furthermore, we selected from Refs. [6,7] data, all those on the high side of (the to be estimated) T_c , in order to avoid the difficulty of the treatment below T_c , but we let T_c to be a free parameter. Thus, the number of treated data points was not always the same depending on the initial choice of T_c .

In Section 2, we outline the method and equations used for fitting, considering a priori similar formulae, but having different arrangements of the parameters which should lead to different correlations during the optimization routine. This is not usually much investigated or discussed and thus we feel it worthwhile to stress a few points. In Section 3, we analyze the fitting results and discuss the possible symmetry of the order parameter. Section 4, serves as a conclusion.

2. Method

When significant measurement errors are present in the independent variable values, here denoted as x_i , as well as in the dependent ones, y_i , it is important to use a fitting method which treats these errors together and does not ignore those in the independent variable, as does the usual least-squares fitting approach. The TbZn data were analyzed with such a bivariate method [5], but, one which did not minimize all residuals simultaneously. Instead, a sequential approach was employed which led to neither accurate least-squares minimization nor to a maximum-likelihood solution. But fitting methods exist which avoid this difficulty and yield estimates of the parameter standard deviations as well as those of the free parameters themselves.

A powerful and convenient method for treating such an “errors-in-variables” situation has recently been discussed and illustrated [10]. It has been installed in the (freely available from the authors) computer program GENLS. This weighted nonlinear least-squares program allows one to use arbitrary individual uncertainties for the x and y data values, s_{x_i} and s_{y_i} . When $s_{x_i} = 0$ for all $i = 1, 2, \dots, N$ data values, the method reduces to ordinary weighted nonlinear least squares. Here, we shall use the GENLS program to fit the $N = 33$ TbZn data values listed in [6,7] for the ca. one degree temperature range (199.5–200.8 K) just above T_c . The program minimizes the following objective function O of the weighted residuals R_{x_i} and R_{y_i} ,

$$O = \sum_{i=1}^N \{ [(x_i - X_i)/s_{x_i}]^2 + [(y_i - Y_i)/s_{y_i}]^2 \} = \sum_{i=1}^N \{ [R_{x_i}]^2 + [R_{y_i}]^2 \}, \quad (1)$$

where the X_i 's and Y_i 's are adjusted values of the variables [10]. The standard deviation of the fit, s_F , is calculated from the final converged value of this weighted sum of squares, $O = O_c$, as $s_F = [O_c/D]^{1/2}$, where $D \equiv N - F$ is the number of degrees of freedom and F is the number of free fitting parameters [10]. When values of s_{x_i} and s_{y_i} are used, which at best take account of the actual expected errors in the data, s_F should be close to unity.

The three singular equations which have been used earlier [5–7] to fit the $\text{TbZn}\rho'$ data will be expressed here in terms of the same adjusted variable ρ' and the adjusted dimensionless temperature variable, $x \equiv T/T_0$, where T_0 is conveniently set to 1 K. Then the magnitude of T_c is given by $x_c \equiv T_c/T_0$. The three equations previously used for the temperature region above T_c are

$$\rho' = \rho + (q/\lambda) [1 - |(x - r)/r|]^{-\lambda}, \quad (2)$$

$$\rho' = \rho + q [|(x - r)/r|]^{-\lambda}, \quad (3)$$

where we recall that we do not consider scaling corrections, and

$$\rho' = \rho + q \ln [|(x - r)|], \quad (4)$$

where $r = x_c$. We have also investigated the utility of fitting with the following three variants of Eq. (4):

$$\rho' = \rho + q \ln [|(x - r)/r|], \quad (4a)$$

$$\rho' = q \ln [p/|(x - r)|], \quad (4b)$$

and

$$\rho' = q \ln [pr/|(x - r)|]. \quad (4c)$$

These equations are clearly all equivalent to Eq. (4) as far as their functional dependence is concerned, so they should all yield exactly the same fits (same residuals). However, the different arrangements of the parameters lead to different correlations between them when they are used to fit the same data set. That arrangement which leads to the smallest correlations is generally the one which yields the smallest parameter standard deviation estimates, the best choice.

In some of the earlier works, the dependent variable was taken as ρ'/ρ_c , where $\rho_c = \rho(T_c)$, instead of ρ' . The p and q parameter values obtained here with the choice of ρ' can be converted to those appropriate for the other choices by dividing them by $\rho_c = 84.4922 \mu\Omega \text{ cm}$, i.e. the value at $T = 199.556 \text{ K} \approx T_c$.

3. Fitting results

Besides obtaining some information on the nature of the order parameter symmetry through the related critical exponents, we are concerned with testing two general hypotheses: first, whether the errors-in-variables fitting approach is needed for the present data, and second, whether one of the fitting models can be reasonably judged as more appropriate than the others. Conclusions will be based on a comparison of fitting values of s_F and on comparisons of parameter relative standard deviation (RSD) estimates. Our main GENLS fitting results are listed in Table 1. For

Table 1
 Results of fitting $d\rho/dT$ vs. T data for TbZn using the GENLS errors-in-variables nonlinear least-squares fitting procedure for three different singular equations with several assumptions for the data uncertainties.

Eq.	No.	N	s_x	s_y	s_F	P_1	P_2	$P_3 \equiv T_c/T_0$	$P_4 \equiv \lambda$
2	33	0		1	8.84×10^{-3}	-0.70 ± 0.74	0.27 ± 0.36	199.613 ± 0.082	0.163 ± 0.246
3	33	0		1	8.84×10^{-3}	0.95 ± 1.04	-1.646 ± 0.312	199.613 ± 0.082	0.163 ± 0.246
4	33	0		1	8.735×10^{-3}	0.2612 ± 0.0031	-0.1139 ± 0.0084	199.556 ± 0.028	—
4	33	0		0.01	0.8735	0.2612 ± 0.0031	-0.1139 ± 0.0085	199.556 ± 0.028	—
4	17L	0		0.01	0.8707	0.2678 ± 0.0062	-0.0970 ± 0.0144	199.591 ± 0.034	—
4	17A	0		0.01	0.8688	0.2621 ± 0.0036	-0.1086 ± 0.0090	199.562 ± 0.029	—
4	33	0.0007		0.01	0.8733	0.2621 ± 0.0031	-0.1139 ± 0.0083	199.556 ± 0.028	—
4	33	0.01		0.01	0.8447	0.2612 ± 0.0034	-0.1141 ± 0.0089	199.555 ± 0.031	—

Notation is defined in text, nd here \equiv indicates that the quantity on the left is an estimate of that on the right.

comparison purposes, some estimated parameter values and their estimated standard deviations have been listed with more significant figures than are justified by the results. The first three fits were carried out using ordinary unweighted (i.e., unity-weighted) nonlinear least squares.

There is no series or renormalization group analysis for the critical exponent λ of ρ' near T_c . From the work of Fisher and Langer [11], it can be expected that the critical exponent characterizing ρ' in magnetic materials like TbZn is the same as that for the specific heat, i.e. α . The specific-heat exponent can be roughly estimated from contour plots given in Ref. [12] or from recent new results.

3.1. Power-law fits

The Eq. (2) results presented in Table 1 show that the data lead to a significant parameter estimate here is only for T_c (i.e., only for parameter r is its RSD, the magnitude of the ratio s_r/r , less than unity). As we shall see from the subsequent fits, even this estimate is quite rough. The situation is a little better for the Eq. (3) fitting, since the new q parameter estimate is statistically significant. But it is clear from the large standard deviation of λ that it cannot be distinguished from zero on a statistical basis, in accordance with earlier analyses [5–7]. If λ was in fact close to zero, this would suggest that the system acts like a three-dimensional (3D) XY system with an ($n = 2$) 3D order parameter according to [12,13]. In fact, $\lambda = 0.014$ when derived from more recent work [14] and using the Josephson equality [3]. Some specialists think that for this 3D XY case α , thus λ could be small and negative [15].

The small values of s_F in lines 1–3 of the table show that the choice $s_y = 1$ is inappropriate for the present data. Although p is extremely uncertain here, when it is held fixed at its converged value and a new fit is carried out, the q , r , and λ parameter estimates remain essentially the same but their estimated standard deviations become 0.084, 0.017, and 0.0096, all of which should be considered as significant values. Thus, although the need to fix p in order to obtain significant estimates for the other parameters is unfortunate and greatly reduces the credibility of these estimates, the results nevertheless suggest that the new λ estimate of 0.1635 ± 0.0096 may not be completely meaningless.

A positive value ca. 0.1 for α or λ can be estimated to correspond to 3D anisotropic (so-called XYZ) magnets [16,17]. The 3D Heisenberg magnet has α near $\frac{1}{8}$. The line of constant $\lambda = 0.16$ ($\approx \frac{1}{6}$) is not shown in Ref. [12] but can be estimated to correspond to a system with an order parameter having a symmetric between zero and unity, the latter like an Ising model. For a three-dimensional Ising-like system, corresponding to an ($n = 1$) 3D order parameter should then be $\lambda = 0.104$ [18]. This 0.104 value derived from high-temperature series expansion agrees better with field-theoretic-method values which yield $\lambda = 0.10$ to 0.11 [9,19].

Before leaving power-law fitting, another physical possibility needs consideration. If quadrupolar as well as dipolar effects were present and important, one would expect

a negative α , i.e. $\lambda = -\frac{1}{3}$ for an order parameter with $n = 4$ in 3D [12]. Thus, it is worthwhile to investigate the case of $\lambda < 0$ (see also the above remark on the 3D XY model). We found that fitting using Eq. (3) (or Eq. (2)) with λ free but constrained to be negative did not converge, even with very large number of iterations. On the other hand, when λ was positive and fixed at $\frac{1}{3}$, convergence was rapid and led to the Eq. (3) estimates $p = 0.106 \pm 0.036$, $q = 0.0656 \pm 0.0069$, and $r = 199.42 \pm 0.04$, with a value of s_F of 8.9×10^{-3} . We see that p is not very well determined here; the value of r is not as plausible as the other estimates of this quantity; and the s_F value is larger than s_F values associated with the other fits. These results suggest that it is unlikely that quadrupole effects are important above T_c for the electrical resistivity behavior of the present material, in contrast to the region below T_c as seen in the specific heat or the magneto-elastic effects [20–22].

Although, all the above results were obtained using ordinary nonlinear least squares, it turns out, as discussed below, that errors-in-variables fitting with plausible non zero values of both s_x and s_y do not lead to smaller estimated standard deviations and is thus an unnecessary complication for the present data.

3.2. Logarithmic-law fits

In view of the small value of estimated standard deviations derived in the previous paragraphs, it is worthwhile to investigate the constrained case, i.e. λ strictly equal to zero, corresponding to a log law (Eq. 4), which has in fact one less parameter available for the fit. The first fit with Eq. (4) shows that although s_F is only slightly reduced compared to the value obtained with Eqs. (2) and (3), all the three free parameters of Eq. (4) are now very well determined by the data. Thus, one cannot depend very much here on s_F values alone in picking an appropriate fitting equation. The next results in the table for Eq. (4) fitting used the weighting choice $s_x = 0$ and $s_y = 0.01$. The value of 0.01 was employed earlier [4] and is relatively consistent with the ρ' experimental accuracy estimate of 1% of Sousa et al. [6] for these data. Although the change of $s_x = 0$ and $s_y = 1$ to $s_x = 0$ and $s_y = 0.01$ for Eq. (4) fitting in lines three and four of the table can only chance the value of s_F proportionately, as shown, the line-3 results are included for direct comparison with those above and the line-4 results for comparison with those below. Notice that using $s_y = 0.01$ leads to s_F values much closer to unity, further confirming this choice as a plausible one. Fits using Eqs. 4(a)–4(c) (not shown), led to s_p values over ten times larger than that for the Eq. (4) fit (line-4 in the table), confirming the latter as the more appropriate choice.

In order to obtain some idea of the stability of estimates obtained with Eq. (4) fitting, further fits were carried out using only the first 17 values above T_c (marked 17L in the table) and for the 17 points starting with the one closest to T_c and omitting the subsequent even points (marked 17A in the table). Although the parameter standard deviation estimates are somewhat larger, the parameter estimates mutually agree to within one of their own standard deviations or better, indicating excellent stability.

3.3. Weighting considerations

Thus far, we have dealt with constant weighting uncertainties. But GENLS allows one to use power-law variable uncertainties. We therefore carried out Eq. (4) fits with $N = 33$ and $s_x = 0$ and $s_y = (Y_i)^z$, where $z = 1$ gives proportional weighting and $z = 0.5$ is appropriate for integer data with Poisson-distributed errors [10]. Fitting with several different values of z showed that best results were obtained as $z \rightarrow 0$, leading to constant unity weighting. This is contrary to the hypothesis introduced earlier following Sousa et al. [4] (private communication) that the precision of the data was likely to be varying over the temperature run. See also the discussion of residuals presented below.

Finally, the last two lines in the table, whose results should be compared to those of the fourth line from the top, show the results of error-in-variables fitting runs using the fixed $s_x = 0.0007$, $s_y = 0.01$ and $s_x = 0.01$, $s_y = 0.01$ uncertainty values. The 0.0007 value, as well as even smaller values, have been employed in the earlier bivariate analysis [4], where discussion of these s_x choices may be found. The next to last line in the table shows that the accurate solution using $s_x = 0.0007$, $s_y = 0.01$ is essentially indistinguishable from that with $s_x = 0$, $s_y = 0.01$. Thus, the likely errors in the temperature measurements are evidently sufficiently small to be completely neglected, allowing ordinary nonlinear least-squares fitting to be used for the present data values.

This result is an a posteriori strong evidence of the excellence of the Sousa et al. data [6]. If the errors in temperature were, in fact, very much greater, so that the choice $s_x = 0.01$, $s_y = 0.01$ were appropriate, one would find the results shown in the last line of the table. Even with this much larger choice for s_x , one sees that the parameter estimates and their estimated standard deviations are scarcely changed.

4. Conclusions

In fitting the present singular equations with the TbZn data, it was found necessary to use very stringent iterative convergence criteria in order to obtain consistent estimates of parameter standard deviations and parameter correlation coefficients. Much less stringent conditions were needed just to obtain consistent parameter estimates. In nonlinear least-squares calculations of this kind, the standard deviation and correction estimates are obtained by linearization around final converged conditions and are associated with the elements of the inverse of the design matrix of the problem [23]. They are thus approximations, and ones which may be particularly poor for the correlations since they are calculated from off-diagonal elements of the matrix. Nevertheless, it is worth quoting the correlations estimated from Eq. (4) fitting and with the choices $s_x = 0$, $s_y = 0.01$, $s_x = 0.0007$, $s_y = 0.01$, and $s_x = 0.01$, $s_y = 0.01$. Let us identify the correlation between the m th and n th parameters as C_{mn} . Then the

C_{12} , C_{13} , and C_{23} correlation estimates were 0.56, -0.74 , and 0.95 ; -0.57 , -0.74 , and 0.95 ; and -0.62 , -0.79 , and 0.95 , respectively. They have the same signs and are comparable in magnitude to the Monte-Carlo values obtained earlier [4]. For the $s_x = 0.0007$, $s_y = 0.01$ weighting choice, the correlation between the x and y residuals was found to be 0.81 .

Fig. 1 shows the actual R_{y_i} residuals obtained with the $s_x = 0$, $s_y = 0.01$ weighting choice compared to the R_{x_i} and R_{y_i} residuals obtained with the $s_x = 0.01$, $s_y = 0.01$ values. Note the relative smallness of the R_{x_i} values even with this much-too-large value for s_x and note also that, except at the low-temperature end of the range, the R_{y_i} residuals are virtually identical for the two weighting choices. Examination of the R_{y_i} values shows no apparent trend of their magnitudes with T . Thus, to a good approximation the data involve errors with constant variance and are thus homoscedastic.

Therefore, the present choice of constant s_x and s_y values is indeed appropriate. No R_{x_i} values are shown for the $s_x = 0.0007$ choice since they are more than ten times smaller than those shown. A cumulative normal probability plot of the $s_x = 0$, $s_y = 0.01$ and R_{y_i} values indicated that, to an excellent approximation, these values were sampled from a normal probability distribution. Therefore from the results in the table that $T_c/T_0 \approx 199.556 \pm 0.028$, one can conclude that in similar replications of the experiment there would be a 68% chance that an estimate of T_c/T_0 would fall in the range 199.53–199.59.

The present analyses do not allow a more precise estimate of T_c/T_0 to be obtained. The estimates of the critical temperature obtained here are, fortunately, consistent

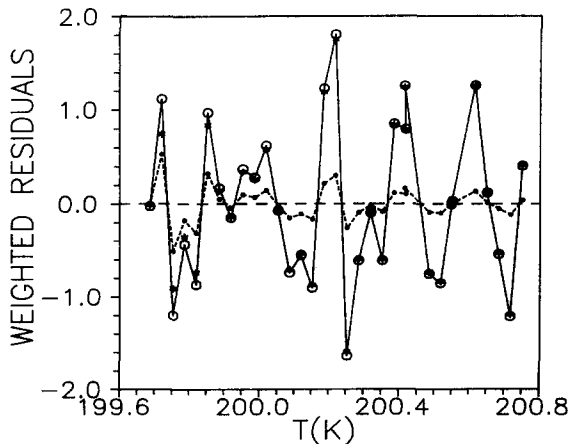


Fig. 1. Weighted residuals, obtained from fits with Eq. (4), vs. unadjusted temperature values. The data uncertainties (s_x , s_y) were $s_x = 0$, $s_y = 0.01$ for the R_y residuals (open circles), and $s_x = 0.01$, $s_y = 0.01$ for the R_y residuals (asterisks) and R_x residuals (bullets). The lines connecting points are included solely to guide the eye.

with those found earlier for the present material. The present work leads, however, to appreciable differences in conclusions about the critical behavior itself.

The above results suggest that it is highly unlikely that λ could be negative and as large as $\frac{1}{3}$, the result expected when quadrupolar as well as dipolar effects are present and important, but they do not entirely rule out the possibility of power-law response with a value of λ near 0.11, the expected value for $\alpha n = 1$, three-dimensional Ising-like model. Nevertheless, it is clear that a log-behavior law, consistent with an $n = 2$ (XY model) for the transition, yields much better defined parameter estimates than does power-law fitting equation with $\lambda > 0$, (even on taking into account some possible lattice softening), unless one of the power-law parameters is held fixed. Even if the log law is not strictly obeyed, the small values of λ theoretically predicted for the 3D XY model are consistent with the log-law predictions. Therefore, we conclude that the 3D XY model is that best representing the phase transition in TbZn, and that well-known [10] fitting methods can lead to reliable conclusions in magnetic systems, just as was the case in previously studied ones like He, near its lambda point. The best estimate for T_c , itself obtained from fitting with the log-behavior law, was 199.56 ± 0.03 K.

It should still be emphasized that, here we investigated only the temperature range above T_c . There is no doubt, that it is of interest to investigate the behavior below a critical temperature if one wants to discuss the critical amplitudes [9,24]. They also help deciding on the order parameter symmetry. We cannot realistically do so here, as we have mentioned in the introduction, first because the contribution from the ordered state is roughly proportional to the square of the magnetization, leading to two extra parameters: the critical exponent and the amplitude of the magnetization; secondly, because in magnetic systems, domains appear in the ordered phase, leading to a definite noise depending on the sweeping (cooling or heating) rate [25]; and because the lattice might have a softening behavior [21,22]. Even though the latter effect is sometimes taken into account in calculating critical exponents, the first two causes make analysis of the low-temperature side uninteresting at our present level.

In conclusion, this study has illustrated difficulties inherent in reaching definitive conclusions about appropriate fitting models and their parameter values near a critical point, even when excellent data are available. Carefully performed statistical data analysis can, however, lead to unambiguous interpretation of physical processes. This was demonstrated here for the (temperature derivative of the) electrical resistivity at a magnetic transition.

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