

# Linear and nonlinear critical slowing down in the kinetic Ising model: High-temperature series

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The difference between the critical exponents ( $\Delta^{(l)}$  and  $\Delta^{(nl)}$ ) of the linear and nonlinear relaxation times of the order parameter ( $\tau^{(l)}$  and  $\tau^{(nl)}$ ) is investigated in the two-dimensional one-spin-flip kinetic Ising model. We have calculated the high-temperature series for  $\tau^{(nl)}$  up to ninth order and made use of the known series for  $\tau^{(l)}$  up to twelfth order. The series are analyzed by the ratio and the Padé-approximant methods. The correlation between the critical-point and critical-exponent estimates in the Padé approximants allows an improvement in the determination of  $\Delta^{(l)}$ . The result,  $\Delta^{(l)} = 2.125 \pm 0.01$ , is higher than previous estimates of  $2.0 \pm 0.05$ . The estimate of  $\Delta^{(nl)}$  is less precise but the result  $\Delta^{(nl)} = 1.95 \pm 0.15$  leads to the conclusion that  $\Delta^{(l)} \neq \Delta^{(nl)}$  and that the difference between the two is of order  $\beta$  (critical index of the order parameter). This is in accord with the scaling prediction.

## I. INTRODUCTION

The application of the renormalization-group formalism to dynamic phenomena<sup>1-3</sup> has greatly improved our understanding of the dynamical aspects of critical behavior. By this method, however, only the singularity of the linear dynamical response has been investigated while experiments<sup>4</sup> and Monte Carlo calculations<sup>5-7</sup> often provide us with the nonlinear response of the system.

It has been noted recently that the critical-point singularities of the linear and nonlinear dynamical responses are different not only in nonergodic<sup>8</sup> but also in ergodic systems.<sup>9</sup> This gives rise to the problem of how to calculate the new set of critical exponents describing nonlinear phenomena.

In the simplest case of purely relaxational systems without conservation laws, scaling arguments<sup>10,11</sup> relate the critical exponent ( $\Delta_Q^{(nl)}$ ) of the nonlinear relaxation time of a physical quantity  $Q$  to the critical exponent ( $\Delta_Q^{(l)}$ ) of the corresponding linear relaxation time by the scaling law

$$\Delta_Q^{(nl)} = \Delta_Q^{(l)} - \beta_Q, \quad (1)$$

where  $\beta_Q$  is the exponent characterizing the scaling of  $Q$  with respect to temperature. For example if  $Q=M$  is the order parameter, then  $\beta_M = \beta$  or if  $Q=E$  is the energy then  $\beta_E = 1 - \alpha$  ( $\beta$  and  $\alpha$  being the critical exponents of the order and heat capacity correspondingly).

The final verification of the scaling prediction (1) is the task of experiments and microscopic theory but some idea about its validity can be obtained from high-temperature expansions and computer simulations. Previous Monte Carlo<sup>5,6</sup> and high-temperature expansion<sup>8</sup> works on the two-dimensional one-spin-flip kinetic Ising model,<sup>12</sup> which is a purely relaxational system without conservation laws, suggested that  $\Delta_M^{(nl)} = \Delta_M^{(l)}$ . In

the two-dimensional Ising model, however  $\beta_M = \beta = 0.125$  is so small that the Monte Carlo calculations with their 5%–10% accuracy cannot distinguish between  $\Delta_M^{(nl)}$  and  $\Delta_M^{(l)}$  if the difference is  $\beta$ . In the case of the high-temperature series<sup>8</sup> only six terms have been calculated and there is an algebraic error at the end of the calculation making all the coefficients, and consequently the ratio estimates of  $\Delta_M^{(nl)}$ , in error.

In this paper we have corrected this error and have calculated up to ninth order the high-temperature series for the nonlinear relaxation time of the magnetization in the square lattice one-spin-flip kinetic Ising model. The resulting series have been analysed by both the ratio and the Padé-approximant method. Parallel to the analysis of the nonlinear relaxation time we reanalyzed the linear relaxation-time series. The recently discovered<sup>13</sup> correlation between the estimates of the critical temperature and critical index in the Padé approximants allowed us to determine  $\Delta_M^{(l)}$  with higher precision; the result  $\Delta_M^{(l)} = 2.125 \pm 0.01$  is different than the previously accepted value,  $\Delta_M^{(l)} = 2.0 \pm 0.05$ . Unfortunately, even this correlation method does not give a very precise answer for  $\Delta_M^{(nl)}$ . From the result  $\Delta_M^{(nl)} = 1.95 \pm 0.15$  one can conclude only that  $\Delta_M^{(nl)} < \Delta_M^{(l)}$  and the difference is of order  $\beta$ .

Section II contains a brief discussion of the kinetic Ising model, the linear and nonlinear relaxation times, and also the results of the high-temperature expansion. In Sec. III we analyze the series. Finally, in the Appendix some details of the calculation of the high-temperature expansion are given.

## II. RELAXATION IN THE KINETIC ISING MODEL

The Ising model which played an essential role in developing the ideas about critical phenomena

has no intrinsic dynamics. One of its simplest dynamical generalizations is the one-spin-flip kinetic Ising model.<sup>12</sup> In this model the interaction of the spins with an infinite heat bath gives rise to spontaneous flips of spins. The probability that the  $i$ th spin flips over  $[w_i(\{\sigma\})]$  in a unit of time depends on the state of the system described by the value of the  $N$  spins  $\{\sigma\} = \{\sigma_1, \dots, \sigma_N\}$ ,  $\sigma_k = \pm 1$ . The transition probabilities are chosen so that the system relaxes to the equilibrium of the standard Ising model. Thus the static properties of the model are well known and the system has no complications other than the diverging lifetimes of fluctuations near the critical point.

Denoting by  $P(\{\sigma\}; t)$  the probability of configuration  $\{\sigma\}$  at a time  $t$ , the dynamics of the above model is described by the following master equation<sup>12</sup>:

$$\tau_0 \frac{d}{dt} P(\{\sigma\}; t) = - \sum_i w_i(\{\sigma\}) P(\{\sigma\}; t) + \sum_i w_i(\{\sigma\}_i) P(\{\sigma\}_i; t), \quad (2)$$

where  $\tau_0$  is just a constant setting the time scale and  $\{\sigma\}_i$  is a configuration differing from  $\{\sigma\}$  by flipping over the  $i$ th spin. The condition that the equilibrium distribution is a steady-state solution of (2) does not determine  $w_i(\{\sigma\})$  uniquely. One of the possible forms used in high-temperature expansions is<sup>14</sup>

$$w_i(\{\sigma\}) = \frac{1}{2} [1 - \sigma_i \tanh(\beta E_i)], \quad (3)$$

where  $2E_i$  is the energy change induced by the flip of the  $i$ th spin. In the case of nearest-neighbor interaction  $E_i = J \sum_j \sigma_j$  where  $J$  is the interaction strength and the sum goes over the nearest neighbors of  $i$ .

Having the solution  $P(\{\sigma\}; t)$  of (2) satisfying the initial condition  $P(\{\sigma\}; 0)$ , the time evolution of the order parameter (magnetization) is completely determined

$$M(t) = \sum_{\{\sigma\}} P(\{\sigma\}; t) M(\{\sigma\}), \quad (4)$$

where the sum is over all possible configurations and  $M(\{\sigma\})$  denotes the value of the magnetization in the configuration  $\{\sigma\}$ .

In the one-spin-flip kinetic Ising model the magnetization is not conserved; its relaxation time  $\tau$  in the high-temperature phase ( $\langle M \rangle = 0$ ) can be defined as<sup>8</sup>

$$\tau = \int_0^\infty \frac{M(t)}{M(0)} dt. \quad (5)$$

This definition is meaningful only if there is a prescription as to how the initial state [with  $M(0)$

$\neq 0$ ] is prepared. If at  $t=0$  the system is in equilibrium with an infinitesimal magnetic field, so that  $M(0)$  is also infinitesimal then (5) is the definition of the linear relaxation time<sup>15</sup> and it can be shown that in ergodic systems it is equal to the relaxation time of the equilibrium fluctuations

$$\tau^{(1)} = \lim_{M(0) \rightarrow 0} \tau = \int_0^\infty \frac{\langle M(t)M(0) \rangle}{\langle M^2 \rangle} dt, \quad (6)$$

where the brackets  $\langle \rangle$  denote the equilibrium average without the field.

On the other hand, if  $M(0)$  is finite then (5) is the relaxation time of the magnetization in nonlinear response<sup>8</sup>

$$\tau^{(nl)} = \tau |_{M(0)=\text{finite}}. \quad (7)$$

Approaching the critical point  $\tau^{(1)}$  and  $\tau^{(nl)}$  will in general diverge with different exponents

$$\tau^{(1)} \sim \nu^{-\Delta(1)}, \quad \tau^{(nl)} \sim \nu^{-\Delta(nl)}, \quad (8)$$

where  $\nu = (T - T_c)/T_c$ ,  $T_c$  being the critical temperature.

These critical exponents can be estimated from the coefficients of the high-temperature series of  $\tau^{(1)}$  and  $\tau^{(nl)}$ . The derivation of the series has been discussed in details<sup>8, 16</sup> for both  $\tau^{(1)}$  and  $\tau^{(nl)}$ . Here we quote only the final result. Introducing the operator

$$L = \sum_i w_i(\{\sigma\})(1 - P_i), \quad (9)$$

where  $P_i$  is the spin-flip operator of the  $i$ th spin

$$P_i f(\{\sigma\}) = f(\{\sigma\}_i), \quad (10)$$

$\tau^{(1)}$  and  $\tau^{(nl)}$  can be written as

$$\tau^{(1)} = \langle M L^{-1} M \rangle / \langle M^2 \rangle \quad (11)$$

and

$$\tau^{(nl)} = \langle L^{-1} M \rangle_i / \langle M \rangle_i, \quad (12)$$

where the brackets  $\langle \rangle_i$  denote the average in the initial ensemble with  $M(0) = \text{finite}$ . For simplicity the initial state is chosen to be the completely ordered state.

The operator  $L$  can be split into a part independent of temperature and parts proportional to the high-temperature variable  $v = \tanh(J/kT)$  and to  $v^3$  (for details see Appendix). By expanding  $L^{-1}$  in powers of  $v$  the problem of calculating (11) and (12) reduces to graph counting on a lattice.

In the case of the square lattice  $\tau^{(1)}$  has been calculated<sup>16</sup> up to  $v^{12}$  while  $\tau^{(nl)}$  has been given<sup>8</sup> erroneously up to  $v^6$ . Our calculation yields the following result for  $\tau^{(nl)}$ :

$$\begin{aligned} \tau^{(nl)} = & 1 + 4v + 16v^2 + \frac{148}{3}v^3 + \frac{416}{3}v^4 \\ & + \frac{10\,444}{27}v^5 + \frac{433\,264}{405}v^6 + \frac{3\,515\,524}{1215}v^7 \\ & + \frac{705\,704\,768}{91\,125}v^8 + \frac{1\,584\,670\,852}{76\,545}v^9 + \dots \end{aligned} \quad (13)$$

Some details of the calculations can be found in the Appendix.

III. ANALYSIS OF THE HIGH-TEMPERATURE SERIES

In this section we apply the ratio<sup>17</sup> and the Padé-approximant<sup>18</sup> methods to estimate  $\Delta^{(nl)}$  from the high-temperature series. Since our main goal is to compare  $\Delta^{(1)}$  and  $\Delta^{(nl)}$ , we reanalyze the series for  $\tau^{(1)}$  parallel to the analysis of the series for  $\tau^{(nl)}$ .

In the Ising model the critical value of the expansion variable  $v_c = \sqrt{2} - 1$  is known so that the ratio method gives an unbiased estimate of the critical exponent. The ratio analysis of  $\tau^{(1)}$  and  $\tau^{(nl)}$  are displayed in Table I.

The estimates of  $\Delta^{(nl)}$  (first column of Table I) do not seem to be settled. There is some oscillation in the values of  $\Delta^{(nl)}$  but there is a growing tendency too. The dangers of guessing the critical exponent when the estimates are monotonic or almost monotonic can be seen for the example of  $\Delta^{(1)}$ . Originally,  $\Delta^{(1)}$  was calculated<sup>16</sup> from the series for  $\tau^{(1)}\langle M^2 \rangle \sim r^{-\Delta^{(1)}-1.75}$ . From the monotonic estimates (second column of Table I) it was concluded that  $\Delta^{(1)} = 2.0 \pm 0.05$ . At the same time the values of  $\Delta^{(1)}$  found from the series for  $\tau^{(1)}$  show a much more regular oscillatory behavior. From the third column of Table I it might be concluded that

TABLE I. Ratio estimates of the critical exponents ( $\Delta^{(1)}$  and  $\Delta^{(nl)}$ ) of the linear and nonlinear relaxation time of the magnetization in the square lattice one-spin-flip kinetic Ising model. In the second column  $\Delta^{(1)}$  was calculated from the series for  $\tau^{(1)}\langle M^2 \rangle \sim (v - v_c)^{-\Delta^{(1)}-1.75}$  while the third column estimates are coming from the series for  $\tau^{(1)}$ .

	$\Delta^{(nl)}$	$\Delta^{(l)}$	$\Delta^{(i)}$
1.	1.657	1.564	1.657
2.	2.314	1.806	2.314
3.	1.831	1.898	2.039
4.	1.657	1.911	2.098
5.	1.777	1.937	2.056
6.	1.873	1.960	2.166
7.	1.842	1.980	2.100
8.	1.869	1.986	2.075
9.	1.966	1.999	2.127
10.		2.012	2.167
11.		2.022	2.122
12.		2.028	2.109

$$2.1 < \Delta^{(1)} < 2.2. \quad (14)$$

Comparing the estimates of  $\Delta^{(1)}$  and  $\Delta^{(nl)}$  one expects that  $\Delta^{(nl)} < \Delta^{(1)}$  and the difference is of order 0.1~0.2.

Trying to improve the estimates of  $\Delta^{(1)}$  and  $\Delta^{(nl)}$  we have applied the other familiar technique of extrapolating finite series, the Padé-approximant method. We have formed Padé approximants to the logarithmic derivative of  $\tau^{(nl)}$ ,  $\tau^{(1)}\langle M^2 \rangle$  and  $\tau^{(1)}$ . The Padé tables of critical point and exponent estimates for the case of  $\tau^{(nl)}$  and  $\tau^{(1)}\langle M^2 \rangle$  are presented in Tables II and III. Conventional analysis of these tables would yield  $\Delta^{(nl)} \approx 1.4$  and  $\Delta^{(1)} + 1.75 \approx 3.75$ . Especially the value of  $\Delta^{(nl)}$  is far off the value one would expect from the ratio estimate.

It has been found<sup>19</sup> however that the better the Padé approximant gives the critical point the better is the estimate of the exponent. Furthermore it was noticed<sup>13</sup> that the plot of the exponent estimate against the critical point estimate is a smooth function which crosses the exact critical point at the exact exponent.

In our case almost all the entries in Tables II and III give critical point values lower than the true value  $v_c \approx 0.4142$ . The simple correlation can be observed from the data that the higher the critical point estimate, the higher the exponent estimate. This means that the conventional analysis underestimates  $\Delta^{(nl)}$  and  $\Delta^{(1)}$ . Plotting the exponent estimates against the critical point estimates (Figs. 1 and 2) one can see that both are smooth functions and a straight-line extrapolation to the critical point yields the following values:

TABLE II. Padé-approximant table of the estimates of the critical point  $v_c \approx 0.4142$  (upper numbers) and the critical exponent  $\Delta^{(nl)}$  (lower numbers, underlined) of the nonlinear relaxation time of the magnetization. Asterisk indicates the presence of a defect in the Padé approximant.

$M \setminus L$	0	1	2	3	4	5
3	0.2996 <u>0.754</u>	0.3753 <u>1.244</u>	0.3999 <u>1.516</u>	0.3966 <u>1.465</u>	0.3907 <u>1.359</u>	0.3948 <u>1.447</u>
4	...	0.4247 <u>2.052</u>	0.3970 <u>1.473</u>	0.4036* <u>1.543</u>	0.3931 <u>1.407</u>	
5	0.4000 <u>1.475</u>	0.3655 <u>0.970</u>	0.3908 <u>1.365</u>	0.3940 <u>1.425</u>		
6	0.3749 <u>1.107</u>	0.3850 <u>1.255</u>	0.3997 <u>1.570</u>			
7	0.3993 <u>1.599</u>	...				
8	0.4252 <u>3.204</u>					

TABLE III. Padé-approximant table of the estimates of the critical point  $v_c \cong 0.4142$  (upper numbers) and the critical index  $\Delta^{(1)+1.75}$  (lower numbers) calculated from the series for  $\tau^{(1)} \langle M^2 \rangle$ . Asterisks indicate the presence of a defect in the particular Padé approximant.

$M \backslash L$	3	4	5	6	7	8
3	0.4012 <u>3.313</u>	0.3104* <u>3.292</u>	...	0.4268 <u>4.786</u>	0.3988 <u>2.845</u>	0.4592 <u>11.048</u>
4	0.4098 <u>3.655</u>	0.4132 <u>3.802</u>	0.4114 <u>3.707</u>	0.4120 <u>3.746</u>	0.4112 <u>3.689</u>	
5	0.4137 <u>3.831</u>	0.4120 <u>3.742</u>	0.4119 <u>3.735</u>	0.4117 <u>3.723</u>		
6	0.4107 <u>3.666</u>	0.4119 <u>3.735</u>	0.4121* <u>3.746</u>			
7	0.4126 <u>3.782</u>	0.4116 <u>3.720</u>				
8	0.4107 <u>3.653</u>					

$$\Delta^{(nl)} = 1.95 \pm 0.15 \tag{15}$$

and

$$\Delta^{(1)} = 2.13 \pm 0.02. \tag{16}$$

The large uncertainty in  $\Delta^{(nl)}$  follows from the relatively poor critical-point estimates. One has to extrapolate quite far to reach the exact critical point. Note that this value of  $\Delta^{(nl)}$  is consistent with the ratio estimate.

$\Delta^{(1)} = 2.13 \pm 0.02$  is different from the conventionally expected  $2.0 \pm 0.05$  and it is consistent with the value given in Eq. (14) from the ratio estimates

for  $\tau^{(1)}$ .

$\Delta^{(1)}$  can also be calculated by forming Padé approximants to the logarithmic derivative of  $\tau^{(1)}$  and again plotting the estimates of  $\Delta^{(1)}$  against the estimates of  $v_c$ . The result is again that of Eq. (16) with about the same accuracy. The accuracy, however, can be increased if one notices that in the Padé approximants there are two unphysical poles on the imaginary axis at about the same distance from the origin as the physical pole. These singularities can be transformed away by the Euler transformation<sup>20</sup>

$$w = 2v / (1 + v/v_c). \tag{17}$$

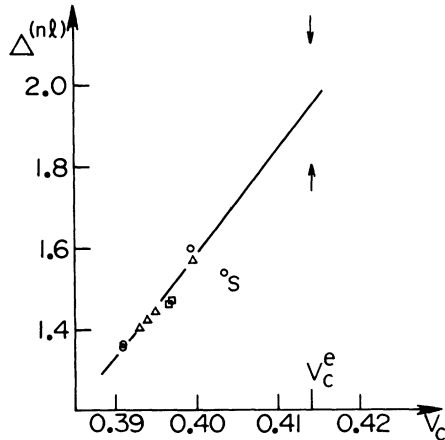


FIG. 1. Correlation between the estimates of the critical exponent of the nonlinear relaxation time  $\Delta^{(nl)}$  and the estimates of the critical point  $v_c$  in Padé approximants calculated by keeping 8 (triangles), 7 (circles), and 6 (squares) terms in the high-temperature series of  $d \ln \tau^{(nl)} / dv$ . The letter *s* denotes the presence of defect in the particular Padé approximant. Straight line extrapolation gives  $\Delta^{(nl)} = 1.95 \pm 0.15$  at the exact value  $v_c^e = \sqrt{2} - 1$ .

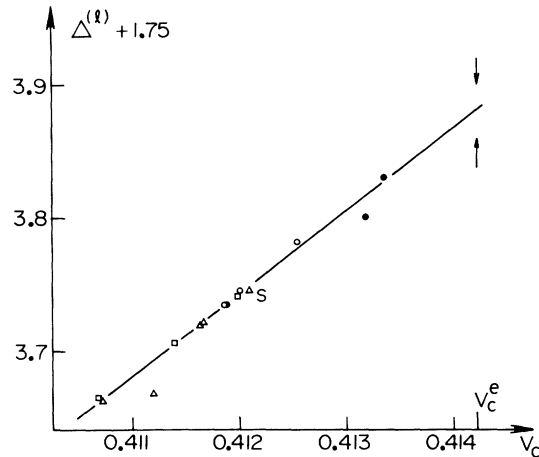


FIG. 2. Padé-approximant estimates of the critical exponent  $\Delta^{(1)+1.75}$  and the critical point  $v_c$  calculated by retaining 11 (triangles), 10 (circles), 9 (squares), and 8 (closed circles) terms in the high-temperature series of  $d \ln (\tau^{(1)} \langle M^2 \rangle) / dv$ . The letter *s* indicates the presence of a defect in the Padé approximant. The straight line extrapolates to  $\Delta^{(1)+1.75} = 3.88 \pm 0.02$  at the exact critical point  $v_c^e$ .

Padé approximants are then formed to  $d \ln \tau^{(1)}/dw$  and analyzed by the previously described correlation method (Fig. 3). We arrive at the estimate

$$\Delta^{(1)} = 2.125 \pm 0.01. \quad (18)$$

Although the analytical structure of  $\tau^{(nl)}$  is similar to that of  $\tau^{(1)}$  transformation like (17) does not lead to improved accuracy in  $\Delta^{(nl)}$ .

We have tried a number of other methods taken from the literature for estimating  $\Delta^{(1)}$ ,  $\Delta^{(nl)}$  and  $\Delta^{(1)} - \Delta^{(nl)}$  but none gave better accuracy than the above correlation method.

The conclusions to be drawn from the above ratio and Padé-approximant analysis are as follows: (i)  $\Delta^{(nl)} < \Delta^{(1)}$  and the difference is of order  $\beta$ . (ii) Since in the two-dimensional Ising model every critical index is a simple fraction from (19) one guesses that  $\Delta^{(1)} = \frac{17}{8}$ . If the scaling law  $\Delta^{(nl)} = \Delta^{(1)} - \beta$  holds then this implies  $\Delta^{(nl)} = 2$ . (iii) The Monte Carlo result<sup>6</sup>  $\Delta^{(1)} = 1.85 \pm 0.1$  is clearly not from the critical region. Since in the temperature region from where this estimate comes the static exponents already assume their asymptotic value this would mean that the dynamical critical region is narrower than the static one. An alternative explanation, of course, is that we do not understand something about Monte Carlo experiments. (iv) The constant  $c$  in the renormalization group calculations [ $\Delta^{(1)} = \nu(2 + c\eta)$ , see Ref. 1] which is independent of dimension near  $d=4$  dimension ( $c \approx 0.73$ ) approaches  $c = \frac{1}{2}$  as  $d \rightarrow 2$ . This is differ-

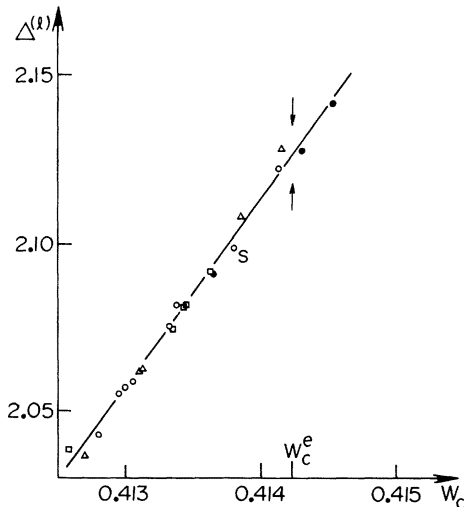


FIG. 3. Padé-approximant estimates of  $\Delta^{(1)}$  and the critical point  $w_c$  after transforming away the unphysical singularities in  $\tau^{(1)}$  by using the Euler transformation  $w = 2\nu/(1 + \nu/v_c)$ . Notation is the same as on Fig. 2. At the exact  $w_c^e = v_c^e = \sqrt{2} - 1$  the critical exponent of the linear relaxation time is  $\Delta^{(1)} = 2.125 \pm 0.01$ .

ent from the many component order parameter case<sup>1</sup> where  $c \rightarrow 0$  as  $d \rightarrow 2$ . If  $c$  is a smoothly varying function of dimension, then one expects that, for  $d=3$ ,  $0.5 < c < 0.73$ . Analyzing the high-temperature series of  $\tau^{(1)}$  in the  $d=3$  kinetic Ising model (Fig. 4) one finds

$$\Delta_3^{(1)} = 1.32 \pm 0.03. \quad (19)$$

It suggests that  $c \approx 1$  although the uncertainties of the static exponents and the uncertainty of  $\Delta_3^{(1)}$  itself allows a range of values  $0.5 \leq c \leq 3$ . From (19) the scaling prediction for  $\Delta_3^{(nl)}$  is  $\Delta_3^{(nl)} \approx 1$ .

Returning to the problem of the scaling law  $\Delta^{(nl)} = \Delta^{(1)} - \beta$ , it would be desirable to have a few more terms in the series of  $\tau^{(nl)}$ . Probably it would be also worthwhile to calculate the series for  $\tau^{(nl)}$  in the three-dimensional kinetic Ising model since the difference between  $\Delta^{(nl)}$  and  $\Delta^{(1)}$  would be much larger ( $\beta \approx 0.31$ ) than in the two-dimensional case.

#### ACKNOWLEDGMENTS

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#### APPENDIX: HIGH-TEMPERATURE EXPANSION OF $\tau^{(nl)}$ IN THE SQUARE LATTICE

The operator  $L$  [Eq. (9)] can be split into three parts<sup>8</sup>  $L = L_a + L_b + L_c$ , where

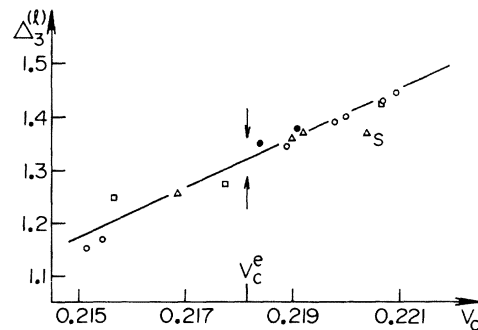


FIG. 4. Correlation between the estimates of the critical point  $v_c$  and the critical exponent  $\Delta_3^{(1)}$  of the linear relaxation time of the magnetization  $\tau_3^{(1)}$  in the cubic lattice one-spin-flip kinetic Ising model calculated from Padé approximants to  $d \ln \tau_3^{(1)}/dv$  retaining 9 (triangles), 8 (circles), 7 (squares), and 6 (closed circles) terms in the high-temperature series. The "exact" value  $v_c^e$  is estimated from the susceptibility series. At  $v_c^e$  the straight-line extrapolation gives  $\Delta_3^{(1)} = 1.32 \pm 0.03$ .

$$L_a = \frac{1}{2} \sum_k (1 - P_k), \quad (\text{A1})$$

$$L_b = \frac{a}{2} \sum_k \sum_{l \in k} \sigma_l \sigma_k (1 - P_k), \quad (\text{A2})$$

$$L_c = \frac{c}{2} \sum_k \sum_{l_1 \in k} \sigma_{l_1} \sigma_{l_2} \sigma_{l_3} \sigma_k (1 - P_k), \quad (\text{A3})$$

where  $P_k$  is the spin-flip operator discussed in Sec. II and  $l \in k$  means that  $l$  is one of the nearest neighbors of the lattice point  $k$ . The coefficients  $a$  and  $c$  have the following expansions in the high-temperature variable  $v = \tanh(J/kT)$ ,

$$\begin{aligned} a &= \frac{1}{8} \tanh(4J/kT) + \frac{1}{4} \tanh(2J/kT) \\ &= v - 3v^3 + 15v^5 - 85v^7 + 493v^9 + \dots \end{aligned} \quad (\text{A4})$$

and

$$\begin{aligned} c &= \frac{1}{8} \tanh(4J/kT) - \frac{1}{4} \tanh(2J/kT) \\ &= -2v^3 + 14v^5 - 84v^7 + 492v^9 + \dots \end{aligned} \quad (\text{A5})$$

In terms of the operators (A1)–(A3)  $\tau^{(nl)}$  (13) can be expressed as

$$\begin{aligned} \tau^{(nl)} &= N^{-1} \sum_{m=0}^{\infty} \left\langle \left[ (L_a - L_b)^{-1} L_c \right]^m \right. \\ &\quad \left. \times (L_a - L_b)^{-1} \sum_i \sigma_i \right\rangle_0, \end{aligned} \quad (\text{A6})$$

where  $\langle \rangle_0$  denotes the average in the completely ordered state.

Noticing that  $L_a M = M$  and  $L_b M = azM$  where  $z$  is the number of nearest neighbors  $\tau^{(nl)}$  can be rewritten in the following form:

$$\begin{aligned} \tau^{(nl)} &= N^{-1} (1 - 4a)^{-1} \\ &\quad \times \sum_{k=0}^{\infty} \left\langle \left[ (L_a - L_b)^{-1} L_c \right]^k \sum_i \sigma_i \right\rangle_0 = \sum_{k=0}^{\infty} a_k. \end{aligned} \quad (\text{A7})$$

Since  $L_c \sim v^3$  the calculation of  $\tau^{(nl)}$  up to  $v^9$  involves only the calculation of this sum up to  $k=3$ . The zeroth-order term is easily found

$$a_0 = (1 - 4a)^{-1}. \quad (\text{A8})$$

After expanding  $(L_a - L_b)^{-1}$  and introducing simplifying notations  $L_a^{-1} L_b = X_b$ ,  $L_a^{-1} L_c = X_c$  and  $N^{-1} (1 - 4a) X_c \sum_i \sigma_i = S$ ,  $a_1$  is expressed as

$$a_1 = \sum_{l=0}^{\infty} \langle X_b^l S \rangle_0, \quad (\text{A9})$$

and since  $X_b \sim v$ ,  $S \sim v^3$  the sum has to be calculated up to  $l=6$ . The results of the graph counting exercise (details of the graph counting problem can be found in Refs. 8 and 16) is

$$\langle S \rangle_0 = 4c/3(1 - 4a), \quad (\text{A10})$$

$$\langle X_b S \rangle_0 = 16ac/3(1 - 4a), \quad (\text{A11})$$

$$\langle X_b^2 S \rangle_0 = 736a^2c/27(1 - 4a), \quad (\text{A12})$$

$$\langle X_b^3 S \rangle_0 = 9920a^3c/81(1 - 4a), \quad (\text{A13})$$

$$\langle X_b^4 S \rangle_0 = 129\,920a^4c/243(1 - 4a), \quad (\text{A14})$$

$$\langle X_b^5 S \rangle_0 = 1\,658\,176a^5c/729(1 - 4a), \quad (\text{A15})$$

$$\langle X_b^6 S \rangle_0 = 20\,873\,216a^6c/2187(1 - 4a). \quad (\text{A16})$$

$a_2$  in (A7) is a double sum

$$a_2 = \sum_{k,l=0}^{\infty} \langle X_b^k X_c X_b^l S \rangle_0 \quad (\text{A17})$$

and up to  $v^9$  only the terms  $k+l \leq 3$  has to be found:

$$\langle X_c S \rangle_0 = 16c^2/5(1 - 4a), \quad (\text{A18})$$

$$\langle X_b X_c S \rangle_0 = 224ac^2/15(1 - 4a), \quad (\text{A19})$$

$$\langle X_c X_b S \rangle_0 = 160ac^2/9(1 - 4a), \quad (\text{A20})$$

$$\langle X_b^2 X_c S \rangle_0 = 237\,376a^2c^2/3375(1 - 4a), \quad (\text{A21})$$

$$\langle X_b X_c X_b S \rangle_0 = 156\,992a^2c^2/2025(1 - 4a), \quad (\text{A22})$$

$$\langle X_c X_b^2 S \rangle_0 = 5\,312a^2c^2/81(1 - 4a), \quad (\text{A23})$$

$$\langle X_b^3 X_c S \rangle_0 = 3\,274\,112a^3c^2/10\,125(1 - 4a), \quad (\text{A24})$$

$$\langle X_b^2 X_c X_b S \rangle_0 = 10\,384\,768a^3c^2/30\,375(1 - 4a), \quad (\text{A25})$$

$$\langle X_b X_c X_b^2 S \rangle_0 = 1\,710\,464a^3c^2/6075(1 - 4a), \quad (\text{A26})$$

$$\langle X_c X_b^3 S \rangle_0 = 324\,928a^3c^2/1215(1 - 4a). \quad (\text{A27})$$

Finally, from  $a_3$  one has to find only one term

$$\langle X_c^2 S \rangle_0 = 19\,808c^3/1575(1 - 4a). \quad (\text{A28})$$

The terms (A9)–(A13) and (A18) have been calculated previously<sup>8</sup> and our result agrees with that calculation. The difference arises after summing up these terms and substituting the expressions (A4) and (A5) for  $a$  and  $c$  in these formulas. We believe the correct answer is given by (13) in Sec. II.

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