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LETTER TO THE EDITOR

Analytic properties of thermodynamic functions at first-order phase transitions

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Abstract. We develop a method of analytic continuation at first-order phase transitions and apply it to the $d = 2$ Ising model in an external field. The method employs a function built of transfer matrix eigenvalues, which provides rapidly convergent approximations to the stable free energy f and its derivatives for all $H \geq 0$. We confirm recent series analysis results on the existence of an essential singularity at $H = 0$. There is also indication of a spinodal line, and $H_{sp}(T) < 0$ is estimated in the range $0 < T \leq 0.6T_c$.

The theory of metastability at first-order phase transitions is an open area in statistical physics. Equilibrium states are described by a well established ensemble theory. However, no such general description of metastable states exists. Attempts to develop a more rigorous theory of metastability (see Penrose and Lebowitz 1979) are confined to relatively simple models. In this Letter we suggest a new method of approximate analytic continuation beyond the coexistence curve, using the eigenvalues of the transfer matrix (TM), and apply it to the $d = 2$ Ising model (a more detailed presentation will be given elsewhere (Privman and Schulman 1982)).

The droplet model (Fisher 1967, Langer 1967) predicts an essential singularity at the coexistence curve. The nature of this singularity has recently been studied by several methods in the $d = 2$ Ising model using TM (Newman and Schulman 1977, McCraw and Schulman 1978), renormalisation group (Klein *et al* 1976) and series analysis methods. Baker and Kim (1980) resummed the low-temperature series and derived an expansion of $m(H)$ (magnetisation) in powers of H . The series is apparently divergent in a way consistent with droplet model predictions. Evidence for the singularity at $H = 0$ was also found in related series analyses (Baxter and Enting 1979, Enting and Baxter 1980).

The foregoing studies find no indication of a spinodal line. Such a spinodal singularity at which $\chi \sim (H - H_{sp}(T))^{-\sigma}$, $0 < \sigma < 1$, has been conjectured on the basis of mean field ($\sigma_{MF} = \frac{1}{2}$) and other phenomenological models (Klein 1981). Existence of a spinodal, its position and properties are a long-standing open problem (Domb 1976). Let us suggest the following heuristic observation. The divergence of χ would imply a divergent correlation length ξ , and in particular at H near H_{sp} ξ would exceed the radius R of the 'critical droplet'. At these H —before reaching H_{sp} —we expect fluctuations to destroy the metastable phase. Therefore we anticipate a smoothed second-order phase transition (as if the system were in a finite volume $\sim R_{H=H_{sp}(T)}^d$). For $\xi \sim R$ there is therefore an enhancement of the droplet decay mechanism which may be interpreted as 'the limit of metastability'. Detailed dynamical arguments

(Langer 1974) suggest a similar picture. Note however that when the spinodal region is approached, the analytically continued free energy (which we consider below) ceases to describe the system ('static' thermodynamic functions cannot be precisely defined (Langer 1974)). It is apparently the case that in the analysis of *finite* power series the spinodal may nevertheless manifest itself as a *bona fide* singularity. Such a situation was probably observed by Gaunt and Baker (1970) and by Ditzian and Kadanoff (1979). In our calculations evidence is found for both the essential singularity and the spinodal.

Consider the $d=2$ Ising model on an $N \times M$ lattice with periodic boundary conditions at fixed $T < T_c$. The energy of the configuration $\{\sigma\}$ of the spins $\sigma_{ij} = \pm 1$ is

$$E\{\sigma\} = - \sum_{ij} \sigma_{ij}(\sigma_{ij+1} + \sigma_{i+1j} + H), \quad i + N \equiv i, j + M \equiv j.$$

The $2^N \times 2^N$ TM is defined between two column configurations σ and σ' ,

$$(TM)_{\sigma\sigma'} = (TM)_{\sigma'\sigma} = \exp\left(\frac{\beta}{2} \sum_{i=1}^N (\sigma_i \sigma_{i+1} + \sigma'_i \sigma'_{i+1} + 2\sigma_i \sigma'_i + H\sigma_i + H\sigma'_i)\right)$$

where $i + N \equiv i$ and $\beta = 1/T$. The free energy per spin of the $N \times \infty$ 'strip' ($M \rightarrow \infty$) is

$$f_1^{(N)} = -(\beta N)^{-1} \log \lambda_1^{(N)},$$

where $\lambda_j^{(N)}$ are the eigenvalues of the TM ($\lambda_1^{(N)} > \lambda_2^{(N)} \geq \lambda_3^{(N)} \geq \dots$). $\lambda_1^{(N)}(H)$ is a branch of an analytic function whose other branches are $\lambda_j^{(N)}$ and whose eigenvectors belong to the completely symmetric representation of the group of symmetry operations of the TM. Henceforth we denote by TM the restriction to the invariant subspace, by $\lambda_1^{(N)} > \lambda_2^{(N)} > \lambda_3^{(N)} > \dots$ the corresponding eigenvalues, and $f_i^{(N)} \equiv -(1/\beta N) \log \lambda_i^{(N)}$. The infinite system stable free energy $f = \lim_{N \rightarrow \infty} f_1^{(N)}$ consists of two branches f_{\pm} (figure 1(a)) with a cusp at $H=0$ (presumably the metastable

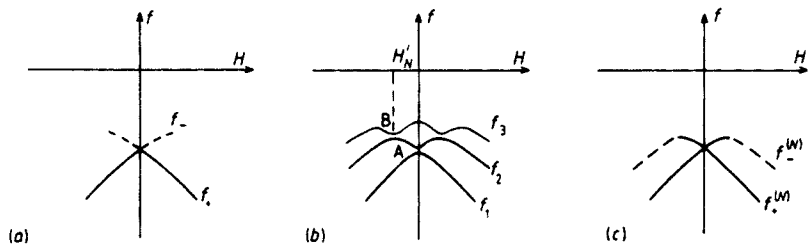


Figure 1. (a) Free energy branches f_{\pm} in the $N \rightarrow \infty$ limit. (b) The lowest free energy branches $f_1^{(N)}$, $f_2^{(N)}$ and $f_3^{(N)}$ (subscript N suppressed); for $|H| \rightarrow \infty$ $f_1 \sim -2 - |H|$, $f_2 \sim (-2 - |H|)(1 - 2/N)$. (c) The modified free energies $f_{\pm}^{(N)}$.

continuations in figure 1(a) have non-zero imaginary parts). The derivative of $f(-m_{\pm}(H))$ has a jump at $H=0$ (twice the spontaneous magnetisation m_s) and the next derivative ($-\chi$) has a delta function contribution at $H=0$. Higher derivatives of f have derivatives of the delta function at $H=0$. This behaviour is approximated by f_1 and its derivatives $D^k f_1^{(N)}$, where $D \equiv \partial/\partial H$. Let us study this property in more detail. The functions $f_1^{(N)}$, $f_2^{(N)}$, $f_3^{(N)}$ are represented schematically in figure 1(b). At

$H=0$ $f_2^{(N)}$ and $f_1^{(N)}$ are asymptotically degenerate with gap size $\Delta^{(N)} = (f_2^{(N)}(0) - f_1^{(N)}(0))/2 \propto e^{-C(T)N}/N$. For the Ising model this is known from Onsager (1944); in general, asymptotic degeneracy as a mechanism for phase transitions has been emphasised by Kac (1966). For $H \gg \text{constant} \times \Delta^{(N)}$, $f_1^{(N)}$ behaves like f_+ ; however in the 'mixing' region, $|H| = O(\Delta^{(N)})$, its magnetisation ($-Df_1^{(N)}$) changes from positive to negative. A continuation with positive magnetisation is along the portion A (figure 1(b)) of $f_2^{(N)}$. At some $H'_N < 0$ (figure 1(b)) $f_2^{(N)}$ encounters higher branches $f_3^{(N)}, f_4^{(N)}, \dots$. For sufficiently low N these higher encounters are separate near-degeneracies (this can be shown in the limit $T \rightarrow 0$, N fixed), and one may continue along the portion B of $f_3^{(N)}$, etc. Such a procedure was suggested by Newman and Schulman (1977) and used by McCraw and Schulman (1978) to obtain numerical estimates of the analytic continuation of f_+ . However, this method runs into difficulty because branch points of the TM eigenvalues do not seem to approach the real $H < 0$ axis in the limit $N \rightarrow \infty$, T fixed. Although the TM provides an analytic continuation, its domain is probably not maximal, and while it presumably gives information about the maximally continued object via dispersion relations (see Schulman *et al* 1978), the continuation apparently does not include the $H < 0$ real axis. The central near-degeneracy at $H = 0$ is however well defined even for large N and governs the behaviour of $f_1^{(N)}$ and $f_2^{(N)}$ for $|H| < |H'_N|$. We assume the following behaviour of H'_N for large N : $H'_N \sim -\text{constant}/N$. This assumption is supported by the exact solution at $H = 0$ (which gives $f_3(0) - f_2(0) \sim 1/N$), and by the observation that the 'interaction' of $f_2^{(N)}$ and higher branches is a typical 'fluctuation' ($\sim 1/N$) effect (in the $T \rightarrow 0$, fixed N limit $H'_N \approx -2/N$). Although $f_1^{(N)} \rightarrow f_+ = f$ for $H \geq 0$ we know that the convergence in the region $|H| = O(\Delta^{(N)})$ is poor; in particular, $D^k f_1^{(N)}$ does not converge to $D^k f_+$ at $H = 0$. For this reason we define a pair of functions

$$f_{\pm}^{(N)}(H) = \frac{1}{2}[f_1^{(N)}(H) + f_2^{(N)}(H)] \pm \left\{ \frac{1}{4}[f_1^{(N)}(H) - f_2^{(N)}(H)]^2 - \Delta^{(N)2} \right\}^{1/2}. \quad (1)$$

By definition of $\Delta^{(N)}$ the argument of the square root is proportional to H^2 near zero and the square root is therefore a single-valued function which is chosen to be proportional to $-H$. Thus on the real H axis we obtain two intersecting branches (figure 1(c)) with

$$f_{\pm}^{(N)} = \frac{1}{2}(f_1^{(N)} + f_2^{(N)}) \mp \text{sgn}(H) \left[\frac{1}{4}(f_1^{(N)} - f_2^{(N)})^2 - \Delta^{(N)2} \right]^{1/2}.$$

It turns out that for $H \geq 0$ $f_+^{(N)}$ is a better approximation to f_+ than $f_1^{(N)}$ in the following two senses: (1) $D^k f_+^{(N)} \rightarrow D^k f_+$ for $H \rightarrow 0^+$, and (2) numerical convergence of the derivatives is much better for positive H away from 0 as well. The motivation for the definition of $f_{\pm}^{(N)}$ is simple. $f_1^{(N)}$ and $f_2^{(N)}$ have a near crossing, a rapid interchange of the form of their eigenvectors and presumably a pair of branch points at $H \approx \pm i\Delta^{(N)}$ where they are exactly degenerate (Newman and Schulman 1977). Moreover, on the scale of $\Delta^{(N)}$ this crossing is distant from any other, so that if one thinks of $f_1^{(N)}$ and $f_2^{(N)}$ as a two-level system, it is reasonable to expect to be able to parametrise them as eigenvalues of a matrix of the form

$$M = \begin{pmatrix} f_+^{(N)} & \Delta^{(N)} \\ \Delta^{(N)} & f_-^{(N)} \end{pmatrix}$$

where the functions $f_+^{(N)}(H) = f_-^{(N)}(-H)$ break the $H \leftrightarrow -H$ symmetry similar to spontaneous breakdown in f_{\pm} . Moreover, if outside the crossing region $f_1^{(N)} \sim f_1^{(N)}(0) - m_1|H| + O(H^2)$, then we expect the functions $f_{\pm}^{(N)}$ to behave like $f_{\pm}^{(N)} \sim f_{\pm}^{(N)}(0) \mp m_{\pm}H + O(H^2)$. By requiring the eigenvalues of M to be $f_1^{(N)}$ and $f_2^{(N)}$ we

obtain equation (1). Let us consider the way in which $f_+^{(N)} \rightarrow f_+$ for $H \geq 0$. For the difference $|D^k f_+^{(N)} - D^k f_+|$ ($H \geq 0$) there are two contributions. First, for $|H| = O(\Delta^{(N)})$ $D^k f_+^{(N)}$ approximates the $-2m_s D^{k-2} \delta(H)$ term in $D^k f$ and performs violent fluctuations of magnitude $\sim [\Delta^{(N)}]^{(1-k)} \sim e^{(k-1)CN} N^{(k-1)}$; using $f_+^{(N)}$ eliminates this problem. Second, finite-size effects occur because one is still using only an $N \times \infty$ lattice. Fisher (1971) has argued that with periodic boundary conditions these effects should be $O(e^{-N/\xi})$ so that we can expect good convergence for $D^k f_+^{(N)}$. We can establish the following properties of $f_+^{(N)}$.

(1) $f_+^{(N)}(H)$ does not have the branch points associated with the $f_1^{(N)} - f_2^{(N)}$ crossing; therefore assuming $f_2^{(N)}$ has its crossing with $f_3^{(N)}$ at a distance of order $1/N$, $f_+^{(N)}(H)$ will be analytic in a circle with radius of order $1/N$.

(2) Using (1), all derivatives $D^k f_+^{(N)}$ at 0 approach the corresponding right derivatives $D^k f(H \rightarrow 0^+) = D^k f_+(0)$ of the exact free energy for $N \rightarrow \infty$.

For proof see Privman and Schulman (1982), but we here mention that to show $D^k f_+^{(N)}(0) \rightarrow D^k f_+(0)$ the following additional property is needed (as a sufficient condition):

$$\lim_{N \rightarrow \infty} [D^k f_+^{(N)}(1/N^{k+2}) - D^k f_+(1/N^{k+2})] = 0.$$

This condition reflects the shrinking of the 'mixing' region faster than any power of $1/N$ (in practice $\Delta^{(N)} \sim e^{-CN}/N$). Note that the modified free energy $f_+^{(N)}$ does not provide analytic continuation of f_+ to $H < 0$. As mentioned above, $f_+^{(N)}$ provides rapidly convergent approximations to all derivatives of f_+ for $H \geq 0$. Calculating a finite number of derivatives at some $H_0 \geq 0$, we obtain a truncated power series for the magnetisation

$$m(H) = 1 + \sum_{k=0}^{\infty} c_k (H - H_0)^k, \quad (2)$$

where the c_k are approximated by $c_k^{(N)} = -(1/k!)(D^{k+1} f_+^{(N)})_{H=H_0} - \delta_{k0}$. The series (2), obtained from the TM, may be used for analytic continuation by conventional series analysis methods.

Consider first the case $H_0 = 0$. We calculated $\lambda_1^{(N)}$ and $\lambda_2^{(N)}$ numerically to about 30 figure accuracy (to avoid round-off errors) for $N = 3, 4, \dots, 9$ at several closely spaced H values. An interpolating polynomial was used to estimate the first 10 derivatives of $f_+^{(N)}$ ($f_+^{(N)}(H=0)$, $c_0^{(N)}, \dots, c_9^{(N)}$). For fixed T this takes about 1.5 hours on the IBM/370 Technion computer. The convergence (with N) is faster for lower temperatures and lower derivatives. With $N \leq 9$, accurate values of all c_0, \dots, c_9 are obtained in the range

$$0 < u \leq 0.3u_c$$

where $u = e^{-4/T}$ is the 'low-temperature' variable ($u_c = 3 - 8^{1/2}$). The temperature $u = 0.1u_c$ is interesting because the coefficients c_1, \dots, c_{24} were calculated by Baker and Kim (1980) using numerical resummation of the low-temperature series. In table 1 we list $f_+^{(N)}(0)$, $c_0^{(N)}, \dots, c_9^{(N)}$ for $N = 7, 8, 9$. Our estimates of the c_n (table 1) are in agreement with the values of Baker and Kim (1980). In figure 2(a) we plot the ratios c_n/c_{n-1} as functions of n for various u values. The apparent linear asymptotic behaviour (figure 2) implies that the radius of convergence of the power series is zero. We checked this linear behaviour for 17 different u values in the range $0.03 \leq u/u_c \leq 0.30$. The nature of the essential singularity at $H = 0$, as implied by the linear c_n/c_{n-1}

Table 1. The coefficients $c_n^{(N)}$ ($m_+^{(N)} = -Df_+^{(N)} = 1 + \sum_{n=0}^{\infty} c_n^{(N)} H^n$) and the values $f_+^{(N)}(0) + 2$ for $u = 0.1u_c$ ($N = 7, 8, 9$). Our estimates are compared with the exact $f(0) + 2$ (Onsager 1944) and c_0 (Yang 1952), and with c_1, \dots, c_9 of Baker and Kim (1980).

	$N = 7$	$N = 8$	$N = 9$	$N \rightarrow \infty$ estimates	Previous results
$f(0) + 2$	-2.999 829 440 001 ...	-2.999 829 442 303 ...	-2.999 829 442 357 ...	-2.999 829 442 35 (± 5)	-2.999 829 442 359 ... $\times 10^{-4}$
c_0	-6.323 422 395 24 ...	-6.323 422 416 22 ...	-6.323 422 416 84 ...	-6.323 422 416 8 (± 6)	-6.323 422 416 86 ... $\times 10^{-4}$
c_1	1.384 959 437 8 ...	1.384 959 467 6 ...	1.384 959 468 7 ...	1.384 959 468 (± 1)	1.384 961 (± 4) $\times 10^{-3}$
c_2	-1.639 627 943 ...	-1.639 628 216 ...	-1.639 628 228 ...	-1.639 628 22 (± 1)	-1.639 628 (± 8) $\times 10^{-3}$
c_3	1.504 764 90 ...	1.504 766 89 ...	1.504 766 99 ...	1.504 767 0 (± 1)	1.504 767 (± 2) $\times 10^{-3}$
c_4	-1.338 894 2 ...	-1.338 906 6 ...	-1.338 907 4 ...	-1.338 907 (± 1)	-1.338 908 (± 3) $\times 10^{-3}$
c_5	1.350 472 1 ...	1.350 542 6 ...	1.350 548 0 ...	1.350 548 (± 6)	1.350 547 (± 7) $\times 10^{-3}$
c_6	-1.631 683 ...	-1.632 055 ...	-1.632 088 ...	-1.632 09 (± 3)	-1.632 092 (± 2) $\times 10^{-3}$
c_7	2.314 43 ...	2.316 28 ...	2.316 48 ...	2.316 5 (± 2)	2.316 502 (± 3) $\times 10^{-3}$
c_8	-3.731 1 ...	-3.739 9 ...	-3.741 0 ...	-3.741 (± 1)	-3.741 159 (± 6) $\times 10^{-3}$
c_9	6.697 0 ...	6.737 2 ...	6.743 3 ...	6.743 (± 6)	6.744 12 (± 1) $\times 10^{-3}$

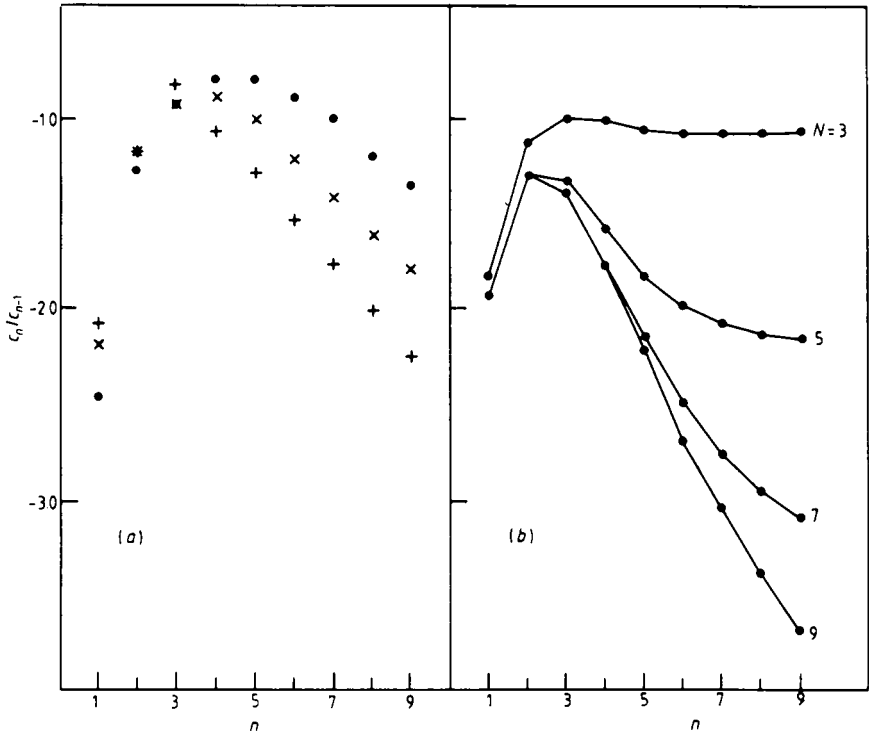


Figure 2. (a) Plot of c_n/c_{n-1} as a function of n for $u=0.05u_c$ (full circles), $u=0.1u_c$ (crosses) and $u=0.15u_c$ (plus symbols). (b) Plot of $c_n^{(N)}/c_{n-1}^{(N)}$ for $u=0.3u_c$, $N=3, 5, 7, 9$.

behaviour, was discussed in detail by Baker and Kim (1980). Lowe and Wallace (1980) showed that it is consistent with field-theoretic droplet model predictions. The approach to straight line behaviour may be followed visually for $u=0.3u_c$, since the convergence is relatively slow in this case. In figure 2(b) we plot the ratios $c_n^{(N)}/c_{n-1}^{(N)}$ for $N=3, 5, 7, 9$. The singularity of f_+ at $H=0$ is built up when the singularities of $f_+^{(N)}(H)$ accumulate along the negative H axis; the nearest ones are at $|H| \propto 1/N$, so that the radius of convergence vanishes like $1/N$ for large N .

Let us now consider the series (2) for non-zero $H_0 > 0$. The motivation is that for $H_0=0$ the coefficients c_n are dominated by the factorially growing contribution from the singularity at zero. To study the structure of $m_+(H)$ for $H < 0$, it is better to use some finite $H_0 > 0$. The coefficients $c_k^{(N)}$ are calculated using the same method. Convergence (with N) improves when H_0 increases; however, longer computer times are needed because $H_0 > 0$ is not a symmetry point. Most results quoted below were obtained with $N \leq 8$ (some with $N \leq 9$). For a fixed temperature $u=0.1u_c$, we performed Padé analysis of the series for χ'/χ ; in a short power series we expect an apparent singularity at $H_{sp}(T)$ with the leading contribution of the form $\chi'/\chi \sim (-\sigma)/(H-H_{sp})$. For $H_0=0, 0.05, 0.10, 0.15, 0.20$ the majority of poles and zeros of different Padé approximants lie on the negative H axis, suggesting a branch cut along the $H < 0$ axis. This trend nearly disappears for $H_0=0.30, 0.40$, presumably because a short power series becomes insensitive to a weak singularity at $H=0$.

In the cases $H_0 = 0.05, 0.10, 0.15$, we clearly observed one stable pole at $H_{sp} = -0.415 \pm 0.015$, whose residues give $\sigma = 0.07 \pm 0.02$. Identification of this pole as a spinodal must be considered with great caution. The spinodal (if it exists) is masked by the essential singularity at $H = 0$, and the branch cut makes the applicability of the Padé method rather questionable and opens the door for systematic errors. The apparently stable pole that we observed may also be an artifact of Padé analysis of series as short as those we used. Support for the identification of this apparent singularity as evidence of a spinodal comes from its reasonable temperature dependence (see below). We sought confirming evidence for a singularity from TM eigenvalues, looking for some dramatic change in gap size (for a gap at approximately fixed negative H and varying T). None was found. Note however, that the value of the apparent critical exponent $\sigma \sim 0.07$ implies that the spinodal is an extremely weak effect in the $d = 2$ Ising model.

For fixed $H_0 = 0.1$ we analysed χ'/χ series for $u/u_c = 0.02, 0.04, \dots, 0.30$. The values of σ from different Padé approximants are spread over the range $0.04 \leq \sigma \leq 0.09$, but individual approximants vary slowly with temperature. The $H_{sp}(u)$ values are summarised in figure 3. Our values are compared with the scaling form of $H_{sp}(T)$

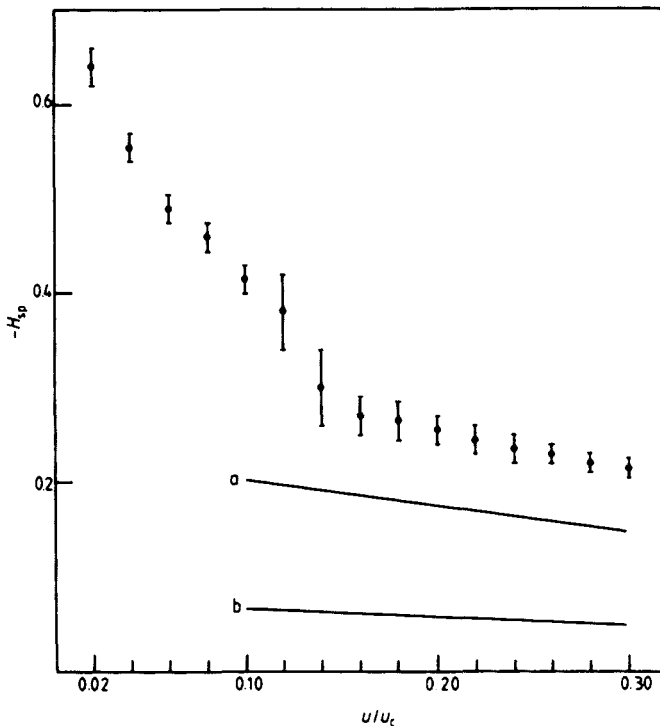


Figure 3. H_{sp} as a function of $u = \exp(-4/T)$ in the temperature range $u \leq 0.3u_c$. The scaling form found by Gaunt and Baker (1970) defines the range between the curves a and b.

for $T \rightarrow T_c^-$. That form gives H_{sp} between the curves a and b in figure 3, namely $\tanh(\beta H_{sp}) \sim (-0.39 \pm 0.20)(1 - T/T_c)^{15/8}$. These values were found by Gaunt and Baker (1970) using the high-temperature series. Our $H_{sp}(T)$ apparently approaches the scaling form (note that $u \leq 0.3u_c$ corresponds to $T \leq 0.6T_c$). In the mean field

case $H_{sp}(T)$ has a finite limit when $T \rightarrow 0$. We studied the low-temperature limit in detail (see Privman and Schulman 1982) and found

$$H_{sp}(T = 0) = -0.82 \pm 0.07.$$

In summary, we have suggested a new method of approximate analytic continuation at first-order phase transitions. Our results for the $d = 2$ Ising model confirm the existence of an essential singularity at $H = 0$ and provide an indication of a spinodal region in the low-temperature range.

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