Carmelo Pisani¹ and Colin J. Thompson¹

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We consider an Ising model with Kac potential $\gamma^{\sigma}K(\gamma |\mathbf{x}|)$ which may have arbitrary sign, and show, following Gates and Penrose, that the free energy in the classical limit $\gamma \to 0+$ can be obtained from a variational principle. When the Fourier transform of the potential has its maximum at $\mathbf{p} = 0$ one recovers the usual mean-field theory of magnetism. When the maximum occurs for $\mathbf{p}_0 \neq 0$, however, one obtains an oscillatory or helicoidal phase in which the magnetization near the critical point oscillates with period $2\pi/|\mathbf{p}_0|$. An example with a potential possessing parameter-dependent oscillations is shown to exhibit crossover phenomena and a multicritical Lifshitz point in the classical limit.

KEY WORDS: Kac potential; mean-field theory; variational principle; helicoidal phase; crossover; Lifshitz point.

1. INTRODUCTION

It is well known that the classical mean-field theories for fluid and magnetic systems can be obtained by considering systems with weak, long-ranged potentials of Kac type $\gamma^d K(\gamma \mathbf{r})$ in d dimensions and taking the limit $\gamma \to 0 +$ after the thermodynamic limit.

Following the pioneering work of $Kac^{(1)}$ and others,^(2,3) Lebowitz and Penrose⁽⁴⁾ considered a system of identical particles subject to a two-body interaction potential of the form

$$\phi(\mathbf{r}) = q(\mathbf{r}) + \gamma^d K(\gamma \mathbf{r}) \tag{1.1}$$

where $q(\mathbf{r})$ represents the short-ranged or hard core component and the second term in (1.1) represents the weak, long-ranged attractive part. Assuming that $K(\mathbf{r})$ is nonnegative, symmetric, and Riemann-integrable over any bounded region of *d*-dimensional space, Lebowitz and Penrose

¹ Mathematics Department, University of Melbourne, Parkville, Victoria 3052, Australia.

were able to recover rigorously the classical van der Waals-Maxwell theory of phase transitions by taking the limit $\gamma \rightarrow 0+$ after the thermodynamic limit.

Gates and Penrose⁽⁵⁾ subsequently generalized this result by relaxing the condition that $K(\mathbf{r})$ be nonnegative and replacing it with the requirement that it be simply bounded. In this way they were able to demonstrate deviations from the van der Waals-Maxwell theory.

The classical Curie-Weiss theory of ferromagnetism can also be obtained⁽⁶⁾ by considering an Ising system of spins $\mu_i = \pm 1$, i = 1, 2, ..., N, on a *d*-dimensional lattice with interaction energy

$$E\{\mu\} = -\sum_{1 \le i < j \le N} \gamma^d K(\gamma \mathbf{r}_{ij}) \,\mu_i \mu_j \tag{1.2}$$

where $K(\mathbf{r})$ satisfies the Lebowitz-Penrose conditions and \mathbf{r}_{ij} denotes the lattice vector between spins *i* and *j*. Again, the classical theory is recovered by taking the limit $\gamma \rightarrow 0+$ after the thermodynamic limit.

Our purpose here is to follow Gates and Penrose and consider Ising spin systems with Kac potentials that are not necessarily nonnegative, but are merely bounded and Riemann-integrable. This allows for competing and oscillatory interactions to occur in the system. Like Gates and Penrose, we note that for certain potentials one gets deviations from standard mean-field theory in which the magnetization is not uniform, but possesses spatial oscillations. Similar behavior was found recently for the spherical model with oscillatory and competing interactions.^(7,8) The Gates–Penrose version of the spherical model was also considered recently by Katz.⁽⁹⁾

In the following section we express the free energy for the Ising model in the classical limit as a variational principle in much the same way as Gates and Penrose did for the gas. The extremal equations in our case, however, are straightforward generalizations of the usual mean-field equations and are much easier to analyze, especially in the critical region, where we show in Section 3 that under fairly general circumstances the magnetization below the critical point undergoes spatial oscillations. A particular case examined in Section 4 is shown also to possess a Lifshitz point corresponding to multicriticality of the paramagnetic, ferromagnetic, and oscillatory or helicoidal phases.

Our results are summarized and discussed in the final section, where we speculate on their relevance to spin glasses and, more generally, to systems with long-range RKKY interactions.

2. THE VARIATIONAL PRINCIPLE

Since in the classical limit the results are essentially dimensionindependent, we consider for simplicity the one-dimensional Ising model of N spins μ_i , i = 1, 2, ..., N, with interaction energy

$$E\{\mu\} = -\sum_{1 \le i < j \le N} \gamma K(\gamma |i-j|) \mu_i \mu_j$$
(2.1)

Assuming only that K(x) is symmetric, bounded, and Riemannintegrable over any bounded subinterval of the real line, we show in the Appendix that in the classical limit ($\gamma \rightarrow 0+$ following the thermodynamic limit) the free energy is given by the variational principle

$$-\beta\psi(\beta) = \max_{\{m(x)\}} \lim_{M \to \infty} f_M\{m(x)\}$$
(2.2)

where the functional f_M is defined by

$$f_{M}\{m(x)\} = (4M)^{-1} \iint_{-M}^{M} \beta K(|x-y|) m(x) m(y) dx dy$$
$$- (2M)^{-1} \int_{-M}^{M} \left(\frac{1+m(x)}{2} \log \frac{1+m(x)}{2} + \frac{1-m(x)}{2} \log \frac{1-m(x)}{2}\right) dx$$
(2.3)

and in (2.2) the maximization with respect to m(x) and the limit M to infinity are interchangeable.

Elementary use of the calculus of variations shows that for finite M, the functional f_M is extremal when m(x) satisfies the condition

$$m(x) = \tanh\left[\beta \int_{-M}^{M} K(x-y) m(y) \, dy\right]$$
(2.4)

which will be recognized as a straightforward and obvious generalization of the standard mean-field equation. In fact, when K(x) is nonnegative, the uniform solution $m(x) = m_0$ (independent of x) of (2.4) maximizes f_M and one easily recovers the classical Curie-Weiss theory.

In general, the solution $m_0(x)$ of (2.4) that maximizes f_M can be interpreted as the (zero-field) magnetization density and when substituted into (2.2) and (2.3) the free energy is given by

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$$-\beta\psi(\beta) = \lim_{M \to \infty} \left\{ -(4M)^{-1} \iint_{-M}^{M} \beta K(x-y) m_0(x) m_0(y) \, dx \, dy + (2M)^{-1} \int_{-M}^{M} \log \left[2 \cosh \beta \int_{-M}^{M} K(x-y) m_0(y) \, dy \right] dx \right\}$$
(2.5)

which also bears close resemblance to the corresponding Curie-Weiss expression.

As in the case of ordinary mean-field theory, it is not difficult to show that in general the trivial solution m(x) = 0, corresponding to zero spontaneous magnetization, maximizes f_M for temperatures T above a certain critical temperature T_c . To see this, we return to (2.3) and expand the logarithmic terms in a Taylor series to obtain the expression

$$f_M\{m(x)\} = \log 2 + (4M)^{-1} \iint_{-M}^{M} \beta K(x-y) m(x) m(y) dx dy$$
$$- \sum_{n=1}^{\infty} (2M)^{-1} \int_{-M}^{M} [m(x)]^{2n} dx/2n(2n-1)$$
(2.6)

where, since |m(x)| < 1, we have used uniform convergence to interchange integration and summation in the second term.

From (2.6), it is obvious that the trivial solution $m_0(x) = 0$ will correspond to a global maximum provided the quadratic term

$$Q = (4M)^{-1} \iint_{-M}^{M} \beta K(x - y) m(x) m(y) dx dy$$
$$- (4M)^{-1} \int_{-M}^{M} [m(x)]^2 dx \qquad (2.7)$$

in (2.6) is negative-definite for all m(x).

In order to simplify (2.7), we form the periodic extensions of m and K and define their complete Fourier series (on $-M \le x \le M$) by

$$m(x) = (2M)^{-1} \sum_{n = -\infty}^{\infty} \tilde{m}(n\pi/M) \exp(in\pi x/M)$$
(2.8)

where the Fourier coefficients \tilde{m} are given by

$$\tilde{m}(p) = \int_{-M}^{M} m(x) e^{-ipx} dx \qquad (2.9)$$

and similarly for K(x) and its Fourier coefficients $\tilde{K}(p)$. Written in this form, (2.8) and (2.9) reduce immediately to Fourier integrals in the limit $M \to \infty$. For finite M, however, substitution of the Fourier series for m and K into (2.7) yields

$$Q = \frac{1}{2} (2M)^{-2} \sum_{n = -\infty}^{\infty} \left[\beta \tilde{K} \left(\frac{n\pi}{M} \right) - 1 \right] \left| \tilde{m} \left(\frac{n\pi}{M} \right) \right|^2$$
(2.10)

It then follows that the trivial solution m(x) = 0 of (2.4), corresponding to a state of zero spontaneous magnetization, maximizes (2.3) when

$$\beta \tilde{K}(n\pi/M) - 1 \leq 0 \quad \text{for all} \quad n \quad (2.11)$$

In particular, if T_c is defined by

$$kT_c = \sup \tilde{K}(p) \tag{2.12}$$

then m(x) = 0 when $T > T_c$.

When $K(x) \ge 0$ the supremum in (2.12) is achieved when p = 0 and T_c takes its Curie-Weiss value

$$kT_{\rm CW} = \tilde{K}(0) = \int_{-\infty}^{\infty} K(x) \, dx \tag{2.13}$$

in the limit $M \to \infty$. In this case m_0 is uniform and nonzero only when $T < T_{CW}$. More generally, when the supremum is reached at some $p_0 \neq 0$,

$$kT_c = \tilde{K}(p_0) > kT_{\rm CW} \tag{2.14}$$

In such a case one might expect a nonzero and nonuniform spontaneous magnetization for $T < T_c$.

In the following section we examine the generalized mean-field equation (2.4) in the critical region just below T_c for $p_0 \neq 0$ and show that an oscillatory solution of (2.4) maximizes $f_M\{m(x)\}$.

3. CRITICAL REGION ANALYSIS

In this section we investigate solutions of (2.4) in the critical region $\beta = \beta_c + \varepsilon$, where $\beta_c = [\tilde{K}(p_0)]^{-1}$, $p_0 \neq 0$, and ε is small and positive. As $\varepsilon \to 0+$, $m(x) \to 0$ by continuity, so we can invert (2.4) and expand in powers of m(x) to obtain

$$(\beta_c + \varepsilon) \int_{-\infty}^{\infty} K(x - y) m(y) dy = m(x) + \frac{1}{3} [m(x)]^3 + \cdots$$
(3.1)

where we have taken $M \to \infty$ in the previous expression for m(x) in accordance with (2.2).

At the critical point ($\varepsilon = 0$) the linear approximation to (3.1) has solutions $m_0 \exp(\pm ip_0 x)$ for arbitrary m_0 . For $\varepsilon > 0$ we retain the cubic term in (3.1) and by symmetry look for solutions of the form

$$m(x) = a\cos(p_0 x) + b\cos(3p_0 x) + \cdots$$
 (3.2)

Direct substitution of (3.2) into (3.1) yields

$$\left(1 + \frac{\varepsilon}{\beta_c}\right) a \cos(p_0 x) + \frac{1}{\beta_1} \left(\beta_c + \varepsilon\right) b \cos(3p_0 x) + \cdots$$

= $\left[a + \frac{1}{4}(a^3 + a^2b + 2ab^2)\right] \cos(p_0 x)$
+ $\left[b + \frac{1}{12}(a^3 + 3b^3 + 6a^2b)\right] \cos(3p_0) + \cdots$ (3.3)

where

$$\beta_1 = [\tilde{K}(3p_0)]^{-1} > \beta_c \tag{3.4}$$

Equating coefficients of $\cos(p_0 x)$ and $\cos(3p_0 x)$, we have that

$$\varepsilon/\beta_c = \frac{1}{4}(a^2 + ab + 2b^2) \tag{3.5}$$

and

$$(\varepsilon/\beta_1 - A)b = \frac{1}{12}(a^3 + 3b^3 + 6a^2b)$$
(3.6)

where

$$A = 1 - \beta_c / \beta_1 > 0 \tag{3.7}$$

To leading order, we then have

$$a \sim 2(\varepsilon/\beta_c)^{1/2}, \qquad b \sim -(2/3A)(\varepsilon/\beta_c)^{3/2}, \qquad \text{as} \quad \varepsilon \to 0+$$
(3.8)

Obviously, one can include further terms in (3.2) and extend the asymptotic expansion obtained above for small $\varepsilon > 0$. It should be noted, however, that this procedure only works when $p_0 \neq 0$. In the ferromagnetic case, for example, where $p_0 = 0$, it will be seen from (3.4) and (3.7) that A = 0 and hence from (3.6) that b, as well as a, is of order $\varepsilon^{1/2}$. Equation (3.2) then reduces to the usual mean-field result.

We note finally from (2.5), (2.6), (2.10), (3.2), and (3.8) that when

 $p_0 \neq 0$ the free energy and spontaneous magnetization in the critical region are given, respectively, to leading order by

$$-\beta \psi \sim \log 2 + [\beta \tilde{K}(p_0) - 1] a^2$$

= log 2 + 2(T_c/T - 1)² as T \rightarrow T_c - (3.9)

and

$$m_0(x) \sim 2(T_c/T - 1)^{1/2} \cos(p_0 x)$$
 as $T \to T_c -$ (3.10)

4. EXAMPLE

A particularly simple example that gives nonuniform behavior is the long-range oscillatory potential

$$K(x) = \exp(-\lambda |x|) \cos \mu x \qquad (\lambda > 0) \tag{4.1}$$

The Fourier transform of K is

$$\tilde{K}(p) = \lambda [\lambda^2 + (p-\mu)^2]^{-1} + \lambda [\lambda^2 + (p+\mu)^2]^{-1}$$
(4.2)

which has its maximum value

$$\beta_c(\lambda)^{-1} = \max \tilde{K}(p) = \tilde{K}(p_0) = \lambda/2\mu [(\lambda^2 + \mu^2)^{1/2} - \mu]$$
(4.3)

when

$$p_0^2 = 2\mu(\lambda^2 + \mu^2)^{1/2} - (\lambda^2 + \mu^2)$$
 provided $\lambda < \mu \sqrt{3}$ (4.4)

To leading order, from (3.10) the spontaneous magnetization has the form

$$m(x) \sim 2(T_c/T - 1)^{1/2} \cos(p_0 x)$$
 as $T \to T_c -$ (4.5)

provided $\lambda < \mu \sqrt{3}$.

When $\lambda > \mu \sqrt{3}$, $\tilde{K}(p)$ has its maximum at p = 0, in which case

$$\beta_c(\lambda)^{-1} = \tilde{K}(0) = 2\lambda/(\lambda^2 + \mu^2)$$

and the spontaneous magnetization has the usual mean-field ferromagnetic behavior

$$m_0 \sim [3(T_c/T - 1)]^{1/2}$$
 as $T \to T_c -$ (4.6)

And of course when $T > T_c(\lambda)$ we have the paramagnetic phase m(x) = 0 for arbitrary λ and μ .

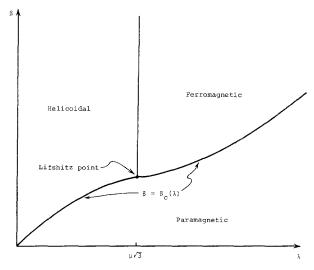


Fig. 1. Phase diagram for the potential $K(x) = \exp(-\lambda |x|) \cos \mu x$.

The situation is summarized in Fig. 1 for some fixed (arbitrary) μ . The region $\lambda < \mu \sqrt{3}$, $\beta > \beta_c(\lambda)$ corresponds to the oscillatory or helicoidal phase, $\lambda > \mu \sqrt{3}$, $\beta > \beta_c(\lambda)$ to the ferromagnetic phase, and $\beta < \beta_c(\lambda)$ to the paramagnetic phase. The meeting point of the three phases at $\lambda = \mu \sqrt{3}$, $\beta = \beta_c(\lambda)$ is usually referred to as a Lifshitz point.

We expect these results to be typical of cases where the potential changes sign. We stress, however, that we have only examined the form of the spontaneous magnetization in the helicoidal region in the neighborhood of the critical point (or curve) $\beta = \beta_c(\lambda)$. It may well be that there are other nonuniform phases and critical lines embedded in this region.

5. DISCUSSION

In this paper we considered a one-dimensional Ising model with a Kac potential $\gamma K(\gamma |x|)$ having arbitrary sign. Following the work of Gates and Penrose, we obtained a variational principle for the free energy in the classical limit $\gamma \rightarrow 0+$ (after the thermodynamic limit). In this formulation the extremal condition is a generalized mean-field equation for the magnetization, which may have nonuniform solutions depending on a spatial variable x.

By examining the mean-field equation in the neighborhood of the critical point we were able to exhibit an oscillatory magnetization solution

with period $2\pi/p_0$ in the case where the Fourier transform of the potential K(x) takes its maximum at $\pm p_0 \neq 0$. When the maximum occurs at $p_0 = 0$ one recovers ordinary mean-field behavior.

An illustrative example was used to show that when the potential has oscillations depending on a parameter one can expect crossover behavior with a Lifshitz multicritical point at the confluence of the helicoidal, ferromagnetic, and paramagnetic phases.

Although we considered only the one-dimensional model, it is clear that the results easily generalize to d dimensions with a Kac potential now of the form $\gamma^d K(\gamma |\mathbf{x}|)$. The only change in the final result is that the integrals appearing are over some d-dimensional bounded domain Ω and are normalized by the volume of this domain [in place of the factors $(2M)^{-1}$ in the one-dimensional case]. Fourier transforms are then taken over R^d and the magnetization in the helicoidal phase has the form

$$m(\mathbf{x}) \sim 2(\varepsilon/\beta_c)^{1/2} \cos(\mathbf{p}_0 \cdot \mathbf{x})$$
(5.1)

where $\mathbf{p}_0 \neq 0$ maximizes the Fourier transform of $K(\mathbf{x})$.

It is interesting to note that in three dimensions the celebrated RKKY potential is in fact a Kac potential of the form considered in this paper with $r = |\mathbf{x}|$,

$$K(r) = r^{-4}(\sin r - r\cos r)$$
 (5.2)

and $\gamma = k_F a$, where k_F is the Fermi wave number and a is the lattice spacing. In this context the limit $\gamma \to 0+$ is usually referred to as the "jellium limit."

For magnetic systems with small $k_{\rm F}a$ (typically $\sim 10^{-1}$) the theory presented here should be directly relevant. One could, in particular, interpret the helicoidal or oscillatory phase for such systems as a kind of spinglass phase. In this respect it would be interesting to attempt a further generalization of the theory to include some degree of randomness.

APPENDIX. DERIVATION OF THE VARIATIONAL PRINCIPLE

For simplicity, we consider a one-dimensional chain of N spins $\mu_i = \pm 1$, i = 1, 2, ..., N, with interaction energy

$$E\{\mu\} = -\sum_{1 \le i < j \le N} \gamma K(\gamma |i-j|) \mu_i \mu_j$$

= $-\sum_{i \ne j=1}^N \gamma K(\gamma |i-j|) \left(\frac{\mu_i + \mu_j}{2}\right)^2 + \frac{1}{2} \sum_{i \ne j=1}^N \gamma K(\gamma |i-j|)$ (A.1)

where, in expressing it in symmetric form, we have used the fact that $\mu_i^2 = 1$.

In order to obtain the variational principle (2.3), we obtain upper and lower bounds on the partition function

$$Z(\beta, \gamma, N) = \sum_{\{\mu\}} \exp(-\beta E\{\mu\})$$
(A.2)

and show that the resulting bounds for the free energy coalesce in the classical limit.

In order to obtain an upper bound on the partition function, we divide the chain of N sites into L strips $\omega_1, \omega_2, ..., \omega_L$ each containing s sites (so that N = Ls). We now define

$$Z(\beta, \gamma, N; m_1, m_2, ..., m_L) = \sum_{\{\mu\}}' \exp(-\beta E\{\mu\})$$
(A.3)

where the sum over $\{\mu\}$ is restricted to configurations with fixed magnetization m_k in strip ω_k , i.e.,

$$m_k = s^{-1} \sum_{i \in \omega_k} \mu_i \tag{A.4}$$

Since each m_k can take only 2s + 1 values, it then follows that

$$Z(\beta, \gamma, N) = \sum_{m_1, \dots, m_L} Z(\beta, \gamma, N; m_1, m_2, \dots, m_L)$$

$$\leq (2s+1)^L \max_{\{m_k, |m_k| \leq 1\}} Z(\beta, \gamma, N; m_1, m_2, \dots, m_L)$$
(A.5)

Now, since

$$\left(\frac{\mu_i + \mu_j}{2}\right)^2 = \begin{cases} 1 & \text{if either } \mu_i = \mu_j = +1 \text{ or } \mu_i = \mu_j = -1 \\ 0 & \text{otherwise} \end{cases}$$
(A.6)

it is not difficult to show that

$$\sum_{\substack{i \in \omega_k \\ j \in \omega_l}} \left(\frac{\mu_i + \mu_j}{2}\right)^2 = \frac{s^2}{2} \left(1 + m_k m_l\right) \tag{A.7}$$

It then follows from (A.1) that the exponent in (A.3) under the restriction (A.4) is bounded above by

$$E^{+}\{m_{k}\} = \frac{\beta\gamma s^{2}}{2} \sum_{k,l=1}^{L} K^{+}(|k-l|)(1+m_{k}m_{l}) - \frac{\beta\gamma}{2} \sum_{i\neq j=1}^{N} \gamma K(\gamma |i-j|)$$
(A.8)

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where

$$K^{+}(|k-l|) = \max_{\substack{i \in \omega_{k} \\ j \in \omega_{l}}} K(\gamma | i-j|)$$
(A.9)

Finally, since there are

$$A(m_k) = s! / [\frac{1}{2}s(1+m_k)!] [\frac{1}{2}s(1-m_k)!]$$
(A.10)

configurations satisfying the restriction (A.4), we obtain from (A.5) and (A.8) the upper bound

$$Z(\beta, \gamma, N) \leq (2s+1)^{L} \max_{\{m_{k}, |m_{k}| \leq 1\}} \left[\prod_{k=1}^{L} A(m_{k}) \right] \exp(E^{+}\{m_{k}\})$$
(A.11)

In a similar way we obtain, for any $m_1, m_2, ..., m_L$, the lower bound

$$Z(\beta, \gamma, N) \ge \left[\prod_{k=1}^{L} A(m_k)\right] \exp(E^{-}\{m_k\})$$
(A.12)

where $E^{-}\{m_k\}$ has the same form as (A.8) but with K^+ replaced by K^- defined by

$$K^{-}(|k-l|) = \min_{\substack{i \in \omega_k \\ j \in \omega_l}} K(\gamma |i-j|)$$
(A.13)

The inequality (A.12) in particular holds for the set of m_k that maximizes the right-hand side of (A.11).

The trick now is to define the (step) function m(x) by

$$m(k\gamma s) = m_k \tag{A.14}$$

and take the "triple Lebowitz-Penrose limit" $L \to \infty$, $\gamma \to 0$, and $s \to \infty$ in an appropriate and convenient fashion. In particular, if we set $L = M/\gamma s$ and hold s fixed, the sums on the right-hand side of the expression (A.8) for $N^{-1}E^+\{m_k\}$ (and similarly for $N^{-1}E^-\{m_k\}$) become Riemann sums (recalling N = Ls) and it is not difficult to see that

$$\lim_{y \to 0+} \lim_{L \to \infty} N^{-1} E^+ \{m_k\}$$

=
$$\lim_{M \to \infty} \beta(2M)^{-1} \iint_0^M K(|x-y|) m(x) m(y) dx dy \qquad (A.15)$$

Similarly, if we use Stirling's formula for the factorial function in (A.10) (for large s), we obtain

$$\lim_{s \to \infty} \lim_{\gamma \to 0+} \lim_{L \to \infty} N^{-1} \log \prod_{k=1}^{L} A(m_k)$$

= $-\lim_{M \to \infty} M^{-1} \int_{0}^{M} \left\{ \frac{1}{2} [1 + m(x)] \log \frac{1}{2} [1 + m(x)] + \frac{1}{2} [1 - m(x)] \log \frac{1}{2} [1 - m(x)] \right\} dx$ (A.16)

Combining these results with the bounds (A.11) and (A.12) for the partition function and noting that

$$\lim_{s \to \infty} \lim_{L \to \infty} N^{-1} \log(2s+1)^L = 0$$
 (A.17)

we obtain, finally, the variational principle (2.3) for the limiting free energy $\psi(\beta)$ defined by

$$-\beta\psi(\beta) = \lim_{s \to \infty} \lim_{\gamma \to 0+} \lim_{L \to \infty} N^{-1} \log Z(\beta, \gamma, N)$$
(A.18)

where, for later convenience, we have used the symmetry of K(x) and defined m(x) = m(-x) for x < 0 to extend the ranges of integration in the above results to the interval (-M, M).

For further discussion and detailed justification of the limiting procedures described above and, in particular, the ordering of the operations in (2.2), the reader is referred to the original work of Gates and Penrose.

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