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A model for re-entrance in antiferromagnetic phases

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Received 8 December 1993

Abstract. A model for antiferromagnetic re-entrant systems is developed. The underlying mechanism for the re-entrance is based on competing interactions between Ising and Potts variables. Renormalization-group calculations are carried out using a modified Migdal–Kadanoff approximation. The resulting global phase diagram is presented and important features are discussed. The model is then applied to liquid crystals, producing qualitative agreement with the behaviour of certain re-entrant liquid crystal systems.

1. Introduction

A phenomenon of considerable interest in statistical mechanics is re-entrance. In a re-entrant system, the usual progression of phases with temperature is reversed, so that a lowering of temperature results in the reappearance of a high-temperature ‘disordered’ phase. The study of re-entrance has a long history, but perhaps the classic example is the re-entrance observed in certain binary liquid mixtures (McEwan 1923, Barker and Fock 1953, Anderson and Wheeler 1978, Walker and Vause 1983). In these systems, the liquid mixture is miscible at high temperature, becomes immiscible as the temperature is lowered, and then re-enters the miscible phase as the temperature is lowered further. Not only is the order of phases novel in such a system, it also appears that the entropy has increased as temperature is lowered—which, in turn, implies thermodynamic instability. Clearly, more is going on than meets the eye. In particular, statistical mechanics is presented with the challenge of answering the following questions: What microscopic interactions are responsible for the re-entrance, and how can the entropy of a system continue to decrease even as it re-enters a—seemingly—less ordered phase? (Goldstein and Walker 1983).

These questions can be addressed most readily with a specific model describing the interactions between various degrees of freedom. For example, a model of interacting Ising and Potts variables, introduced by Walker and Vause (WV) (Walker and Vause 1983), describes re-entrance in binary liquid mixtures. In this model, the Ising variables (Ising 1925) represent the two components of the mixture, whereas the Potts (Potts 1952) variables describe the relative orientations of the molecules—if the relative orientation is correct the molecules hydrogen bond, otherwise no bond is formed. The mechanism for re-entrance, in this case, is that the favourable energy of hydrogen bonding produces a low-temperature miscible phase, while, at the same time, the low orientational entropy associated with the hydrogen bond more than offsets the increase in entropy due to the system becoming miscible again.

In this paper, we study a new type of re-entrant phase transition to be found in the WV model. All previous studies of the model have dealt with re-entrance in the ferromagnetic Ising transition (i.e. miscible to immiscible), which occur in a particular

subspace of the global model. We show that the full WV model displays not only this well studied ferromagnetic re-entrance, but a complementary, and unstudied, antiferromagnetic re-entrance as well. Though the antiferromagnetic transition has no particular relevance to liquid mixtures, it may apply to other physical systems. For example, certain liquid crystals show the following doubly re-entrant sequence of phases with decreasing temperature; nematic, smectic A (bilayer), nematic, smectic A₁ (monolayer) (Cladis 1975, Cladis *et al* 1978). In these systems, the smectic A phase is characterized by molecules that are anti-aligned; hence, the antiferromagnetic Ising transition may provide at least a *qualitative* understanding of the re-entrance mechanism.

Other models have been quite successful in describing such systems (Berker and Walker 1981, Indekeu *et al* 1987). These models are based on a different mechanism from the one studied here—a mechanism in which frustration is the cause of re-entrance. One of the interesting aspects of the re-entrance we present is the complete absence of frustration in our model.

In what follows, then, we describe the general WV model in section 2, with special attention given to the subspace that contains the antiferromagnetic re-entrance. For ease of reference to previous work, we use the original notation wherever possible. Section 3 describes a Migdal–Kadanoff (MK) (Migdal 1975, Kadanoff 1976) renormalization-group calculation, which extends and generalizes the original work on the model. We present global phase diagrams in section 4, and conclude with a brief discussion of the qualitative connection between the model and liquid crystals in section 5.

2. Model

The WV model describes the interactions between Ising variables ($s_i = \pm 1$, denoting two types of molecules, A or B) and Potts variables ($\sigma_i = 1, 2, 3, \dots, q$, representing molecular orientations) appropriate to a hydrogen-bonding liquid mixture. It is given by

$$H = \sum_{\langle i,j \rangle} [K_1(1 - \delta_{s_i s_j})\delta_{\sigma_i \sigma_j} + K_2(1 - \delta_{s_i s_j})(1 - \delta_{\sigma_i \sigma_j}) + K_3\delta_{s_i s_j}\delta_{\sigma_i \sigma_j} + K_4\delta_{s_i s_j}(1 - \delta_{\sigma_i \sigma_j})] \quad (1)$$

where H is the reduced Hamiltonian $-H/k_B T$, with H the lattice Hamiltonian, k_B the Boltzmann constant, T the absolute temperature, and K_n ($n = 1, 2, 3, 4$) the reduced coupling constants described below. In what follows, $K_4 \equiv 0$ is chosen as a reference level. Previous renormalization-group studies of this model (Walker and Vause 1983) have involved only the subspace where $K_3 = 0$, yielding re-entrance in the ferromagnetic phase. Here we focus on a previously unmapped region where $K_3 \neq 0$ and $K_1 = K_2 = K$, which again yields re-entrance, this time in the antiferromagnetic phase. In this subspace, equation (1) becomes

$$H = \sum_{\langle i,j \rangle} [K(1 - \delta_{s_i s_j}) + K_3\delta_{s_i s_j}\delta_{\sigma_i \sigma_j}]. \quad (2)$$

Since only K_3 is associated with Potts variables, we refer to K_3 as the Potts interaction. Similarly, K will be referred to as the Ising interaction. The resulting reduced energy level diagram is shown in figure 1 (note that positive values correspond to favoured interactions). The K_3 , K , and zero levels have degeneracies of $2q$, $2q^2$, and $2q(q - 1)$, respectively, and correspond to interactions in which nearest neighbours are like molecules with equal Potts

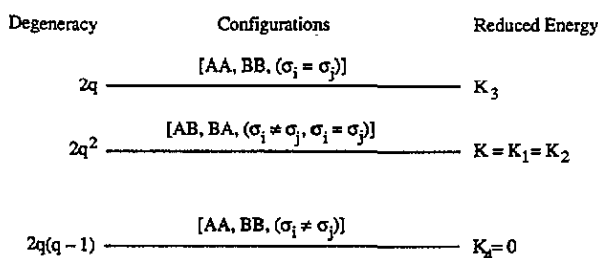


Figure 1. Reduced energy level diagram for $K_1 = K_2$. The sign and magnitude of the interactions is appropriate for re-entrance. Note that the interaction that is energetically most favourable (i.e. most positive) is also the interaction with the lowest associated entropy.

variables, unlike molecules with any Potts variables, and like molecules with unequal Potts variables, respectively.

Several special cases will now be explored (refer to figure 1):

(i) Ising model: ($K_3 = K_4 = 0$). For this case like molecules have the same energy for all σ , i.e. the Potts variables have been decoupled. The reduced Hamiltonian is

$$H = \sum_{(i,j)} K(1 - \delta_{\sigma_i, \sigma_j}) \tag{3}$$

which is a standard Ising model except that the two energy levels, K and zero, have degeneracies of $2q^2$ rather than 2. The extra overall degeneracy of q^2 simply adds a constant to the free energy and so does not alter the critical properties of the system. Therefore, two symmetrically located critical points are expected at $K = \pm K_c$. Since for $K > 0$ unlike-nearest-neighbour occupation is the most energetically favourable configuration, $+K_c$ is an antiferromagnetic critical point. Similarly for $K < 0$ like-nearest-neighbour occupation is the most favourable configuration, therefore $-K_c$ is a ferromagnetic critical point. When $|K| < |K_c|$ the system is in the high-temperature disordered phase.

(ii) $2q$ -state Potts model: ($K = K_4 = 0$). Again there are two energy levels; one level is K_3 with a degeneracy of $2q$ and the other is zero with a degeneracy of $2q(2q - 1)$. The reduced Hamiltonian is

$$H = \sum_{(i,j)} K_3 \delta_{r_i, r_j} \tag{4}$$

with $r_i = 1, 2, 3, \dots, 2q$. This is a $2q$ -state Potts model, therefore only a ferromagnetic critical point at some $K_3 > 0$ is expected for $q > 1$ since the antiferromagnetic Potts model yields no phase transition.

(iii) q -state Potts model: ($K \rightarrow -\infty$). If $K \rightarrow -\infty$, then the K_3 and K_4 levels predominate in the system and the K level can be ignored. Now there are two energies with degeneracies of $2q$ and $2q(q-1)$; a q -state Potts model with an extra overall degeneracy of 2. Note that letting $K \rightarrow -\infty$ means that $s_i = s_j$ for all nearest neighbours, hence the reduced Hamiltonian becomes

$$H = \sum_{(i,j)} K_3 \delta_{\sigma_i, \sigma_j} \tag{5}$$

Again, only a ferromagnetic critical point is expected at some $K_3 > 0$.

In this model, re-entrance occurs due to the dynamic modification of Ising interactions by Potts variables. The system minimizes its free energy $F \equiv E - TS$ at high temperature by being in the state of greatest entropy, the disordered phase, and at low temperature by being in the most energetically favoured state, the ferromagnetic phase described by K_3 where nearest neighbours have equal Potts variables. At intermediate temperatures, however, K_3 has little effect on the system due to its low degeneracy, hence low associated entropy, therefore the ferromagnetic phase is disfavoured. Despite the fact that the antiferromagnetic phase described by K is less energetically favourable, its greater entropy allows the free energy to be minimized as the temperature is decreased from the high-temperature region. (Note that if K_3 is ignored, K describes only this antiferromagnetic transition.) At still lower temperature, before reaching the ferromagnetic transition, the competing Ising and Potts interactions effectively cancel each other out, resulting in the system's re-entrance into the disordered phase.

3. Renormalization-group calculations

The steps for performing the renormalization-group calculations for this model are the same as described in the wv paper (Walker and Vause 1983); that is, we carry out a Migdal-Kadanoff (Migdal 1975, Kadanoff 1976) approximation consisting of an approximate bond-shifting step followed by an exact step which sums out a fraction of the degrees of freedom from the restructured lattice. After bond shifting, the interaction between nearest-neighbour sites i and j is given by

$$\tilde{H}(s_i, s_j, \sigma_i, \sigma_j) = b^{d-1} H(s_i, s_j, \sigma_i, \sigma_j) \quad (6)$$

where b is the length-rescaling factor and d is the dimensionality of the lattice. In order to make direct comparisons with the wv calculations, we adopt their assignment of $d-1 = 1.2$ for the exponent of b . This choice ensures that the MK approximation is in optimum agreement with known results for the three-dimensional Ising model.

Next, we perform an exact decimation on the lattice. We use $b = 3$, which allows antiferromagnetic and ferromagnetic occupations to be treated on an equal footing. The resulting recursion relations are as follows:

$$K'_0 = \ln Z_0 \quad (7a)$$

$$K' = \ln(Z/Z_0) \quad (7b)$$

$$K'_3 = \ln(Z_3/Z_0) \quad (7c)$$

where K'_0 contributes to the free energy. The partial partition functions Z_0 ($s_i = s_j$; $\sigma_i \neq \sigma_j$), Z ($s_i \neq s_j$; σ_i, σ_j arbitrary), and Z_3 ($s_i = s_j$; $\sigma_i = \sigma_j$) are given by

$$\begin{aligned} Z_0 &= q^2 - 3q + 3 + 3q(q-1)x^2 + 3qzx^2 + 3(q-2)z + 3z^2 \\ Z &= 3(q-1)^2x + 6(q-1)zx + 3z^2x + q^2x^3 \\ Z_3 &= (q-1)(q-2) + 3q(q-1)x^2 + 3qzx^2 + 3(q-1)z + z^3 \end{aligned} \quad (8)$$

with $x = \exp(b^{1.2}K)$ and $z = \exp(b^{1.2}K_3)$.

We now discuss several invariant subspaces of our renormalization-group transformation and point out the connections to the special cases of the model discussed in section 2. First, if $K_3 = 0$, then the resulting partial partition functions (8) are

$$\begin{aligned} Z_0 &= Z_3 = q^2(1 + 3x^2) \\ Z &= q^2(3x + x^3). \end{aligned} \tag{9}$$

As a result, we see from equation (7c) that $K'_3 = 0$, hence $K_3 = 0$ is an invariant subspace. Furthermore, $K_3 = 0$ is the Ising subspace, and, as expected, the recursion relation (7b) for K' is independent of q since both Z and Z_3 are proportional to q^2 . If instead we let $K = 0$, we find

$$\begin{aligned} Z_0 = Z &= (2q)^2 - 3(2q) + 3 + 3(2q - 2)z + 3z^2 \\ Z_3 &= (2q)^2 - 3(2q) + 2 + 3(2q - 1)z + z^3. \end{aligned} \tag{10}$$

Clearly, $K' = 0$ for this case, hence $K = 0$ is an invariant subspace. Referring to special case (ii) in section 2, we note that this subspace corresponds to the $2q$ -state Potts model. Similarly, if we let $K \rightarrow -\infty$ then

$$\begin{aligned} Z_0 &= q^2 - 3q + 3 + 3(q - 2)z + 3z^2 \\ Z &\rightarrow 0 \\ Z_3 &= q^2 - 3q + 2 + 3(q - 1)z + z^3 \end{aligned} \tag{11}$$

resulting in $K' \rightarrow -\infty$, hence $K \rightarrow -\infty$ is also an invariant subspace. From special case (iii) in section 2, this limit is the q -state Potts model. Note that the recursion relation for K'_3 is the same for the last two cases, except that q occurs in place of $2q$ in the $K \rightarrow -\infty$ limit, thus showing that the renormalization-group transformation faithfully reproduces the q - and $2q$ -state Potts limits of the original model.

4. Global phase diagram

In this section we explore the global phase diagram of our model in (K, K_3) space. Since this space is invariant (as is easily verified), all of our results can be displayed conveniently in two-dimensional figures. The phase diagram is obtained by successive iteration of the recursion relations given in the previous section. Any initial point, (K, K_3) , will be transformed to a new point (K', K'_3) by the MK transformation, and similarly, each additional iteration maps the current point to another renormalized point. In this sense, a renormalization-group 'flow' is created. Each point in (K, K_3) space flows to a fixed point—that is, a point where further iterations no longer have an effect—and each fixed point can be interpreted as corresponding to a critical point or a given phase of the system. In this way the phase diagram is mapped out into various two-dimensional regions, corresponding to phases, separated by lines of phase transitions, as shown in figure 2. We begin our study by considering the five fixed points displayed in figure 2.

Perhaps the most trivial fixed point is HT, which is located at the origin. That HT is a fixed point is readily verified by noting that if $K = K_3 = 0$, then all the partition functions reduce to $4q^2$. As a result, it is clear from the recursion relations that $K' = K'_3 = 0$. HT is found to be irrelevant in both the K and the K_3 directions, which simply means that it

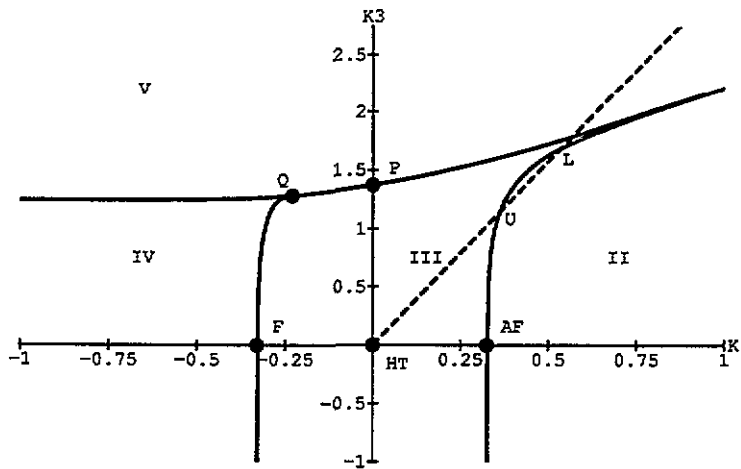


Figure 2. Phase diagram in (K, K_3) space. The diagram shows the following five fixed points: HT, high-temperature sink; AF, antiferromagnetic Ising transition; F, ferromagnetic Ising transition; P, ferromagnetic $2q$ -Potts transition; and Q, multicritical point. In addition, four phases are indicated in the diagram: II, antiferromagnetic Ising; III, disordered; IV, ferromagnetic Ising; and V, ferromagnetic $2q$ -Potts.

attracts flows from a two-dimensional region, labelled III in figure 2. The physical meaning of HT becomes clear when one recalls that the reduced couplings, K and K_3 , are of the form (energy)/ $k_B T$. Thus, if the energies in the system are finite, it follows that $K = K_3 = 0$ corresponds to an infinitely high temperature (HT). Thus, we see that HT is a fixed point corresponding to the disordered phase of the system, and hence, every point in region III represents a disordered state of the system.

Next, we find two mirror-image fixed points in the Ising subspace, $K_3 = 0$. These fixed points, labelled F and AF, are relevant in the K direction and irrelevant in the K_3 direction, meaning that they have one-dimensional domains of attraction extending in the K_3 direction. These domains of attraction are indicated by the lines extending from the points F and AF in figure 2. Since these lines separate regions of difference phases, F and AF are fixed points representing phase transitions. In particular, F and AF control the ferromagnetic and antiferromagnetic Ising transitions, respectively.

A similar fixed point occurs in the $2q$ -state Potts subspace, $K = 0$. In this case we find a fixed point at P which is relevant in the K_3 direction and irrelevant in the K direction. As a result, P has a one-dimensional domain of attraction extending in the K direction, as shown in figure 2. Physically, P represents the ferromagnetic $2q$ -state Potts transition. Since the Potts model with many states has no antiferromagnetic transition, there is no mirror-image point to P; in fact, no fixed points occur on the negative K_3 axis.

The fifth fixed point in figure 2 is the point labelled Q. The first notable feature of this fixed point is that it does not occur on one of the invariant subspaces of the model. Instead, it simply occurs where two phase transitions happen to merge. In addition, Q is relevant in both directions, hence its domain of attraction is restricted to the point itself. Since Q represents the merger of two different phase transitions, we identify it as a multicritical point. Finally, we note that the fixed points P and Q are the $K_3 \neq 0$ versions of P and Q in the original WV model.

Finally, a sixth fixed point should be mentioned, though it does not appear in figure 2. This fixed point governs the line extending to the left of Q, and is located at $K \rightarrow -\infty$.

We find this fixed point to be relevant in the K_3 direction, and to represent a ferromagnetic q -state Potts transition.

In addition to the fixed points, we note that figure 2 shows three types of phase transitions, distinguished by which variables take on long-range order as the transition is crossed. The transitions can be classified as follows: Ising variables order, Potts variables unaffected; Potts variables order, Ising variables unaffected; Ising and Potts variables order simultaneously. For example, the line extending from the fixed point AF is the locus of points where Ising variables assume long-range antiferromagnetic order, while the Potts variables remain disordered. The line passing through the point F is similar, except in this case the long-range order is ferromagnetic Ising.

Potts variables are involved in the remaining two transitions. For example, the line extending to the left of Q is a q -state Potts transition; below this line the Potts variables are disordered, above they have long-range ferromagnetic order. Crossing the line extending to the right of Q, and passing through P, causes all $2q$ variables to order simultaneously. Notice that a thin sliver of disordered phase always exists between the AF and Q-P transition lines.

The existence of antiferromagnetic re-entrance is indicated by the dashed line extending from the origin in figure 2. This line represents a locus of initial conditions for a particular system, with temperature decreasing with increasing distance from the origin. To see this, recall that all the reduced couplings are of the form (energy)/ $k_B T$, as mentioned above. The energy referred to here is an energy of interaction between neighbouring molecules, which one expects to be temperature independent. Therefore, the ratio K_3/K , for example, is a temperature-independent ratio of energies characteristic of the system being studied. The constancy of K_3/K is inherent in the dashed straight line; the fact that the dashed line intersects the AF line twice means the system displays two critical points as a function of temperature.

In particular, at high temperature the system is disordered. As it is cooled, an Ising critical point is encountered at U (denoting an upper critical point). For a range of temperatures below U the system maintains antiferromagnetic Ising order, but as temperature is lowered further to L, a second (lower) Ising critical point occurs. Immediately below this temperature the system is disordered again. At even lower temperatures a third phase transition is observed, this time causing the ordering of all $2q$ variables.

The disordered phase below the point L deserves special attention. Though the system is disordered there, it is substantially more complex than in the region near the origin. To see this, recall that the Ising and Potts interactions compete: Ising interactions favour antiferromagnetic alignment of nearest neighbours, Potts interactions favour ferromagnetic alignment. In the high-temperature disordered phase all interactions are weak, but in the low-temperature disordered phase the interactions are strong—the absence of order in this case is due to the competing interactions effectively cancelling one another. Thus, correlations are strong when the system re-enters the disordered phase. In fact, the increased correlations between Potts variables serves to lower the entropy more than enough to offset the increase in entropy caused by the system re-entering the disordered phase, as discussed above.

Finally, note that if K_3/K is increased—increasing the slope of the dashed line—the temperature range of the antiferromagnetic phase decreases. In fact, for large enough K_3/K the system never enters the antiferromagnetic phase at all, but remains disordered until all $2q$ variables order at a still lower temperature. This is in accord with the previous qualitative discussion; namely, if the Potts interaction, K_3 , is increased enough it can overcome the entropy disadvantage associated with that interaction. As a result, the Potts interactions

out-compete the Ising interactions, and prevent long-range antiferromagnetic order from occurring.

5. Liquid crystal example

We now apply our model for re-entrant systems to liquid crystals. In this example, we focus on molecular species with characteristic polar heads and hydrocarbon tails (Cladis 1975). Only temperature regions below the isotropic–nematic phase transition, in which molecules lie along a preferred axis, are considered. We assume that the liquid volume is made up of cubical molecular-sized cells, each containing an A or B molecule with a polar head pointing in the positive or negative direction ($s_i = \pm 1$), respectively. The interpretation of the reduced energy level diagram of figure 1 is shown in figure 3. The Potts variables ($\sigma_i = 1, 2, 3, \dots, q$) now represent hydrocarbon tail configurations, rather than molecular orientation. For like-directed nearest neighbours, we assume that molecules with equal σ -values have interlocking, and thus interacting tails, while those with different σ -values have non-interacting tails.

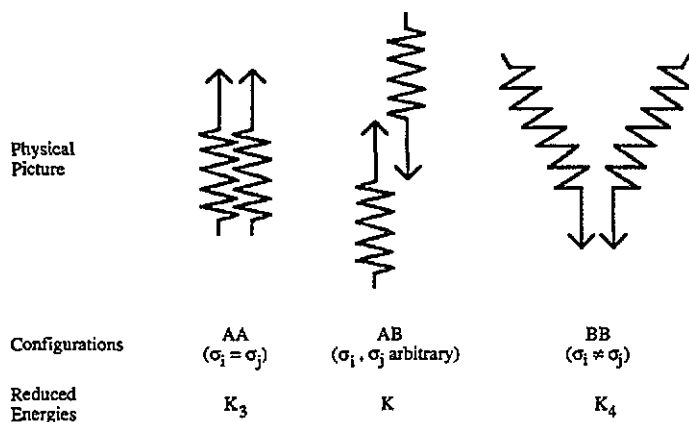


Figure 3. Liquid crystal molecular configurations corresponding to the reduced energy level diagram of figure 1. In this case, *A* refers to an upward pointing molecule, *B* to a downward pointing molecule. The tail configurations are represented by the Potts variables.

The physical portion of the phase diagram in figure 2 is the first quadrant where $K > 0$ and $K_3 > 0$. That K is positive is clear from figure 3, since anti-alignment gives a favourable dipole interaction, and with this alignment tails do not interact. For K_3 we assume that the tail interaction is strong enough to overcome the dipole interaction, making the overall interaction favourable. For our liquid crystal model, phase II denotes a smectic A phase in which unlike-directed molecules are nearest neighbours, phase III is a nematic disordered phase, and phase V is a smectic A_1 phase in which like-directed molecules have interacting tails.

As discussed in the previous section, we can assign a ratio $R = K_3/K$ in order to characterize a particular system. Figure 4 shows the phase diagram plotted as reduced temperature ($T = 1/K$) versus R . The dashed line represents an arbitrarily chosen system with $R = 3.30$. Qualitatively, the diagram correctly describes the behaviour of certain

liquid crystals, such as 8OCB (Lushington *et al* 1980). The system exists in a nematic phase (III) at high temperature, then exhibits a phase transition to the smectic A phase (II) as the temperature is lowered, followed by re-entrance into the nematic phase at still lower temperature, followed by a final transition into the smectic A₁ phase (V).

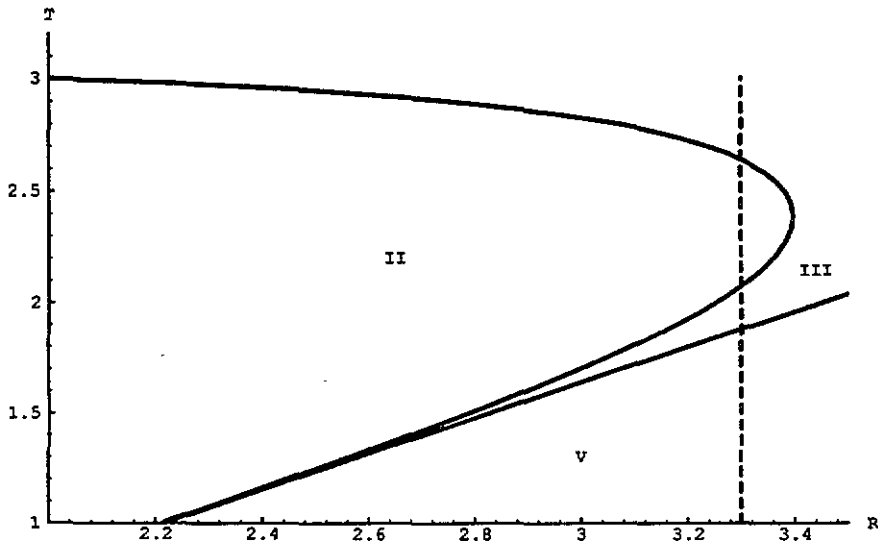


Figure 4. Phase diagram plotted as reduced temperature ($T = l/K$) versus $R = K_3/K$. The dashed line indicates the locus of initial conditions for an arbitrarily chosen value of $R = 3.30$. The numbering of the phases follows that of figure 2.

The number of possible tail configurations q can be changed to alter the shape of the phase diagram. Figure 5 shows how the antiferromagnetic phase boundary of figure 2 becomes taller and steeper as q is increased from $q = 500$ (a) to $q = 50\,000$ (b). In the figure, T_l denotes the lower phase transition temperature, while T_u denotes the upper transition temperature. The dashed lines each correspond to the same value of T_l/T_u so that each intersected curve spans the same relative temperature interval. The resulting T versus R diagram shown in figure 6 reveals the modification in antiferromagnetic phase boundary shape when q increases from $q = 500$ (a) to $q = 50\,000$ (b). This diagram exemplifies how the symmetry of the phase boundary 'knee' can be adjusted to best model a particular system by increasing or decreasing the value of q accordingly.

6. Summary

The study presented here has focused on a previously unexplored subspace of the WV model. In this subspace we find several phase transitions of various types, including an antiferromagnetic Ising transition that exhibits re-entrance. The underlying mechanism of the antiferromagnetic re-entrance—competition between the *energetically* favourable Potts interactions and the *entropically* favourable Ising interactions—is basically the same as that responsible for the ferromagnetic re-entrance observed in binary liquid mixtures. We propose that the antiferromagnetic re-entrance reported here may play a role in the re-entrant nematic–smectic A–nematic–smectic A₁ phase transitions of certain liquid crystals.

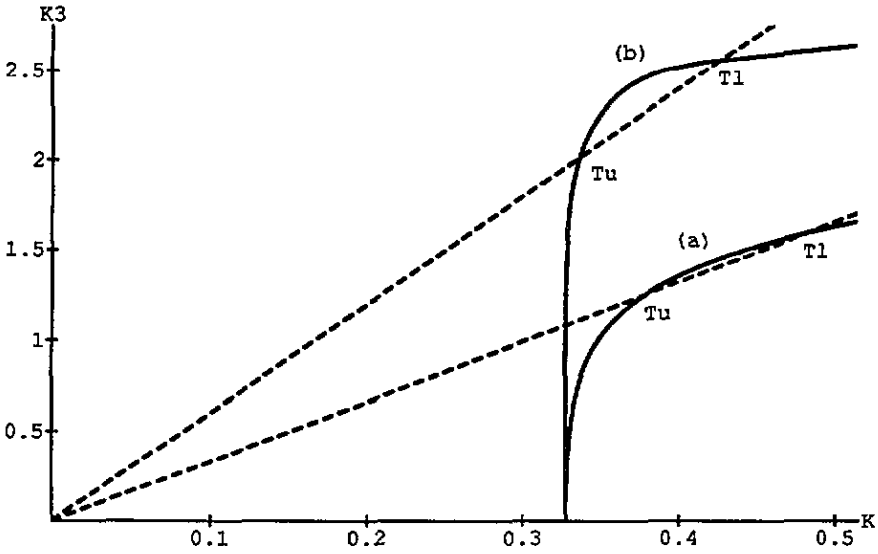


Figure 5. Antiferromagnetic phase boundaries in (K_3, K) space for two different values of q ; $q = 500$ (a) and $q = 50\,000$ (b). Note the increased sharpness of the phase boundary 'knee' as q is increased. T_l and T_u denote the lower and upper phase transition temperatures, respectively. The two dashed lines correspond to different loci of initial conditions for two different values of R ; $R = 3.30$ (a) and $R = 5.97$ (b). These values of R ensure that each line has the same ratio of lower and upper temperatures, $T_l/T_u = 0.78$. This allows phase boundary shapes to be compared, as in figure 6.

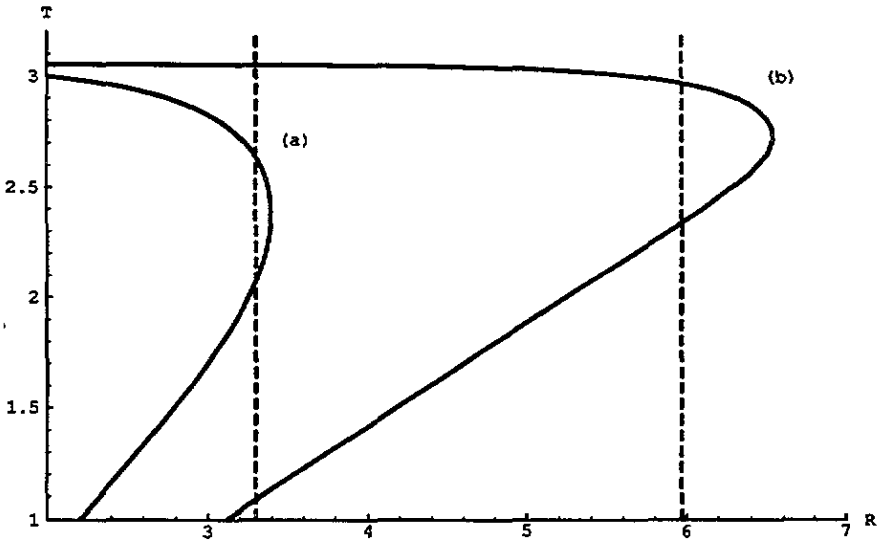


Figure 6. Antiferromagnetic phase boundaries of figure 5 plotted as reduced temperature T versus R . Note the increased sharpness and change in symmetry near the phase boundary apex as q is increased from $q = 500$ (a) to $q = 50\,000$ (b).

Finally, our study has been carried out using a variation of the Migdal-Kadanoff position-space renormalization-group approximation. The transformation we derive here successfully preserves the important qualitative features of the model. Furthermore, the same transformation applied to binary liquid mixtures has proven to give quantitative agreement with experiment.

Acknowledgments

The authors would like to thank Q Li and Y-H Huang for helpful discussions. S M Wood acknowledges the support of the Northwest College and University Association for Science (NORCUS) Graduate Program.

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