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Onsager, Ice, Biomembranes, Dimer Models and the F-Model

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Personal recollections give a sampling of some of Onsager's later interests in ice and biomembranes. The author's involvement in these topics led to modeling a particular biomembrane phase transition using dimer models. Recent work is described for a particularly rich dimer model which is isomorphic to the F-model in three kinds of fields (direct, staggered, and quarter). New results for the full three-dimensional phase diagram show (1) how the anomalous OK multicritical point is destroyed by a direct field, and (2) how a new line of critical points must be added to the phase diagram in direct and staggered fields obtained previously by Baxter, due to diverging susceptibilities in the quarter field.

KEY WORDS: Phase transitions; F-model; dimer models; ice; biomembranes.

1. RECOLLECTIONS

As a young physics graduate student in 1962 I was fascinated by the mysteries of statistical mechanics. After searching unsuccessfully for a statistical mechanician on the Yale physics faculty, I heard about this chemist. Being a snobby young mathematical physicist, I thought it unlikely that a chemist could do deep mathematical science. Nevertheless, I made my way to Onsager, who talked about several possible projects, though I did not really understand all of them until years later. It was clear, however, that his math was deep enough for me. Since three of his advanced physics students had recently finished superconductivity projects, going beyond BCS by including electron correlations, I thought it logical

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to carry on that project and I saw a way to use a computer to do the algebra that would allow carrying the perturbation expansions further. That, however, did not appeal to Onsager, who did not have a high regard for computers. Perhaps by way of explanation, Gell-Mann and Brueckner had used a computer to approximate, replete with error bars, the value of an integral involved in electron correlation functions. Onsager obtained the exact value, which lay embarassingly outside the estimated errors.(1) However, when I visited Lars in Miami a year or so before his death, he was very enthusiastic with his Hewlett-Packard reverse Polish calculator to help do his calculations on ion transport in solution. He impressed me with knowing how to use his a lot better than I knew how to use mine, so I think he would eventually have come to appreciate computers better.

One time in early 1963 when I was talking to Onsager in his office about superconductivity, he closed his eyes and took a little nap. I got the message and stopped working on superconductivity. I might mention, though, that he had a habit of sleeping in seminars, even exceptionally good ones. This did not prevent him from engaging in deep discussion at the end. I also think that Lars preferred expressing himself nonverbally or with very few words. Misguided graduate students thought that this was due to a lack of proficiency with English and some even called his statistical mechanics courses by the names Norwegian I and Norwegian II. But his command of English was exceptional, as evidenced by his habit of doing the *New York Times* crossword puzzles, which were too challenging for most of us. His papers were also written with great verbal skill, though some readers complain about his preference for leaving out many of the more pedestrian mathematical steps. Notice that I say his preference for leaving out pedestrian steps, not his habit of leaving them out. I think that it was a well-thought-out preference because he himself enjoyed working things out himself instead of seeing them all written out. I think he felt that this was the best way to learn and, in turn, this accounted for his method of teaching and communicating.

After awhile I was told by one of Onsager's chemistry graduate students, Kelly Runnels, about a mathematical problem involving the residual entropy of common ice. Onsager had proved that Linus Pauling's calculation was only a lower bound.(2) After I had made a little progress, I talked to Lars about it. The level of his enthusiasm was very great indeed, considerably higher than for superconductivity. The only problem I had with working on ice was explaining to my physics department why it was worthy of a physicist. When I asked Lars for a reason, he simply said that "There is a lot of it around." Statistical mechanicians now appreciate ice as the prototypical vertex model and there are now elegant exact solutions in two dimensions due to Lieb for the six-vertex model(3) and Baxter for the eight-vertex model,(4) though my refined approximation is still the best available for real three-dimensional ice.(5)

A few years after I had obtained my Ph.D. from Lars, I decided that I should branch out from esoteric statistical mechanics and felt that my background in hydrogen-bonded materials such as ice might be useful in biology. When I mentioned this to Lars, he suggested that I visit Yale and work with him on the problem of ion conduction through the membranes of nerve axons. He had a theory that small cations such as sodium might be transported along chains of hydrogen bonds formed from the side chains of proteins. By the way, Onsager did not like to publish much, as is well known. He reserved his publications for his best ideas, and he seldom published things that did not turn out right and did not have high significance. This ion channel idea was one that he slipped into four publications,(6–9) including his Nobel Prize speech. Lars clearly felt that biology was the next great frontier and I think he felt his work on ice to be one route to biology. It is noteworthy that, during the late 1960s and early 1970s when critical phenomena and phase transitions became fashionable, Onsager did not participate, but was thinking about ice and biology. I like to interpret this as his recognition that critical phenomena were in good hands and that he could explore and help initiate another wave of theoretical science.

However, returning to the particular project on ion channel conduction through membranes, new data came in which made us realize that chains of hydrogen bonds could not, in fact, provide enough current for the sodium channels in nerve axions and Onsager dropped this project. Before I mention the sequel to this topic, however, I think it is worth mentioning that Lars did *not* apply his classical irreversible thermodynamics to ion conduction through membranes. The reason is that the energies involved are too large compared to kT, so that one is probably not in the linear transport regime. For more details I refer to one of my papers,(10) which gives references to Onsager's brief remarks on this.

Here is the sequel to the story of Onsager's idea that chains of hydrogen bonds between the side chains of proteins would provide a natural pathway for ion movement through biomembranes. Although such chains would facilitate small ion conduction such as sodium by solvating the ion, they would be much better conduction pathways for protons. Lars was very well aware of this from his ice studies, which focused on protonic semiconductivity. However, in 1969 few people thought proton conduction through membranes was important. That changed rapidly by the mid-1970s with experimental proof of Peter Mitchell's chemiosmotic hypothesis. Neither Lars nor I had noticed this development, but Harold Morowitz at Yale had and Harold remembered something of Lars' earlier ideas. Harold, fearing he would not understand Lars, contacted me and I worked on it with Harold for a bit until we had enough to write a letter to Lars. Sadly, shortly after we mailed the letter, Lars died and the opportunity for an exciting collaboration was lost. I would like to report that this idea swept the bioenergetics field and vindicated Lars' basic idea. That is still premature. We have developed the idea in different contexts(11,12) and it has received a fair hearing. It is being tested experimentally for various membrane proteins, which is not an easy undertaking. My guess is that it will hold up to some extent, probably involving bound water as well as protein side chains to form the proton channels.

I cannot help but tell one more Onsager anecdote that involves the fields of membrane biology and ice. Lars and I were attending a session on membranes at a New York Academy of Sciences meeting in 1971. I was getting a bit bored and was sneaking toward the exit when I looked back and saw Lars practically running after me. He preceeded to tell me about bovulline and drew some pictures. What little I remembered of Latin made me think that bovulline must have something to do with cows, and that fit with the context of a biology meeting. However, after asking a lot of elementary questions, which was the only way to communicate with Lars, I discovered that he had made a much more subtle mental jump. Bovulline is an organic molecule that undergoes multiple sequential tautamerization that performs a permutation of its carbon atoms, as indicated in Fig. 1. The graph of the permutations has order three and rather long shortest cycles with 12 steps before returning to the original state without trivial backtracking. This feature is similar to the graph of the classical basis states in ice as an excess proton performs a random walk, as indicated in Fig. 1. Lars had earlier that year produced a quantum mechanical calculation for the energy density of these ionic states that was exact for a Bethe lattice which has order three and infinitely long shortest cycles. (The actual ice lattice has shortest cycles of 18 steps and 2D ice has shortest cycles of 14 steps.) We had wondered if there is a specific graph-theoretic algorithm that would, in the appropriate limit, produce a Bethe lattice. Lars was suggesting that the graph of permutations of bovulline, suitably generalized to hyperbovulline by adding more carbons to the three legs of the molecule, might do this job. After working on this for awhile back home, I proved that Lars' generalization of bovulline did not do it, but has shortest cycles with 12 steps even in the limit. So, in the best Onsager tradition, this was put into a filing cabinet drawer along with many neat Onsager ideas that did not have maximum significance.

I might mention, however, that the quantum mechanical work on proton mobility in ice(13) did finally get written after I badgered him into doing it. The order of the authors is rather amusing, by the way. Mou-shan

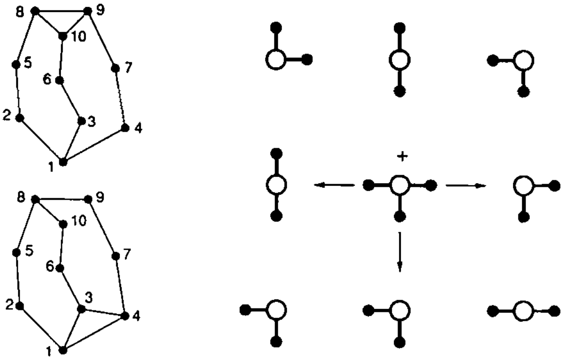

Fig. 1. Left: Two $C_{10}H_{16}$ bovulline molecules related by breaking one of the three bonds in the tight triangle and forming another tight triangle at the other end of the molecule, thereby permuting the carbon numbering that generates a graph of local coordination number 3. Right: One of the basis states of two-dimensional ice with one (+) ionic defect that is connected to three other basis states by shifting an excess proton along one of the three arrows.

Chen did only one of the more minor sections in this paper, but as a student he had brilliantly corrected an Onsager mistake on ionic transport in solution, work that dated back to the 1930s. Onsager published very few mistakes in any of his many fields, so it was much to Chen's credit to find one. Lars, however, procrastinated helping Mou-shan write a paper, but not in telling other researchers about it, until others finally published the correction. So, first authorship on the unrelated proton mobility papers was Chen's reward! Jill Bonner discovered, using matrix methods she had used when working with Michael Fisher, that the quantum mechanical problem could be solved on a Cayley tree and I simplified the analysis. The density of states is amusing, in that it is, in mathematical language, everywhere dense, but nowhere continuous. I pulled all the equilibrium calculations together, wrote it up, and supposed that this would be a satisfactory first paper. But Lars then proceeded, over a period of a year or so, to do the nontrivial transport calculations, based on the Cayley tree calculations rather than on his Bethe approximation. It is a paper that I learned much from and one that I am proud to have been an author of, with many theoretical innovations and relevance to a real material. However, if he had not felt an obligation to his coauthors, I think he would not have published it because it was not a definitive paper. The experimental data were and still are murky, and there is a plethora of other likely choices for the basic transport model in addition to tunneling.

Many of us have wondered how many such excellent, but not outstanding, pieces of work were left in the filing cabinet. Incidentally, I might mention two other items that I know were in that filing cabinet. One was his letter to Bruria Kaufman showing how to obtain the critical exponent β for the Ising model. The second was some scraps of paper from which he could reconstruct a way to checkmate with a bishop and knight—in Kriegspiel (blindfolded chess)! But do not ask me to reproduce the procedure!

Before finishing this set of recollections, let me emphasize that Lars was also involved with many other problems and a number of other students in these later years. Unfortunately, we students did not have the breadth of knowledge to be able to benefit from each other's work with Lars; it was difficult enough to catch up to him in one's own thesis area! I should also emphasize that this activity level was really quite high, but he did not go out of his way to give seminars, so that even his faculty colleagues tended to have only partial pictures that suggested to some that his activity level was lower than it actually was.

2. DIMER MODELS FOR BIOMEMBRANES

Fortunately for me, the same year 1970 when we determined that hydrogen-bonded chains were not appropriate for sodium channels in nerve, I found another biology problem involving membranes and statistical mechanics that I continue to work on today. Although Lars was not involved with this problem, the statistical mechanical aspect is a grandchild of the two-dimensional Ising model that Lars solved first.(14)

The biological connection is different from the ionic transport through biomembranes. The latter problem focused on the proteins embedded in lipid bilayers. The lipid bilayers, however, undergo phase transitions that in turn impact the activity of the membrane proteins. Another focus, therefore, is the lipid bilayer.

The main lipid bilayer phase transition is driven by the conformational melting of the hydrocarbon chains. This is similar to the melting transition in hydrocarbon crystals such as alkanes or polyethylene, with one important difference. Both above and below the lipid bilayer transition one end of each hydrocarbon chain is pinned to a nearly planar interface with water. This imposes an anisotropic boundary condition which is correlated to a considerable difference in the thermal behavior of exactly solvable lattice models.(15)

The simplest model for the main lipid bilayer phase transition allows chains to proceed away from the water interface with a choice of three options that may be modeled as one *trans* and two *gauche* continuations. When this is imposed onto a two-dimensional lattice, it becomes isomorphic to a dimer model, which I have called the K-model, originally

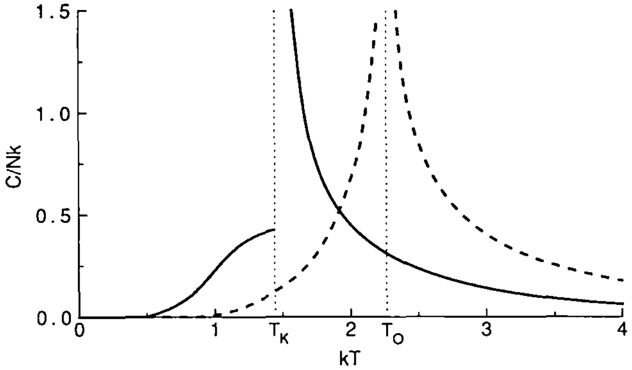

Fig. 2. Specific heat of an O-type transition (dashed lines) compared to a K-type transition (solid lines).

solved by Kasteleyn.(16) The model has also been generalized to allow for a volume increase at the main transition. These developments were recently reviewed,(17) so details will not be given here. It is worth mentioning, however, that despite obvious simplifications required to obtain exactly solvable models, respectable quantitative agreement with experiment has been achieved.(18)

From the point of view of statistical mechanics, the remarkable feature of the K-model and the other models for lipid bilayers is that the phase transition is a totally different type from the phase transition in the two-dimensional Ising model, despite the fact that both models can be solved by the same Pfaffian technique.(16) This is emphasized by the specific heat curves shown in Fig. 2. It is my suggestion that these two types of transition be called O-type and K-type transitions, after Onsager and Kasteleyn, who first obtained them from exact solutions. The traditional alternative to O-type transition is to call this an Ising transition. Also, the K-type transition is better known to many condensed matter physicists as the Pokrovsky–Talapov transition. Considerable detail regarding the features that make a dimer model have transitions of O-type versus K-type can be found in our review.(17)

3. NEW RESULT FOR F-MODEL

Until very recently, all the known phase transitions obtained by the Pfaffian method were either K-type or O-type. Furthermore, even though some models had more than one transition, they were always of the same type unless the basic energy parameters in the models were changed. It had

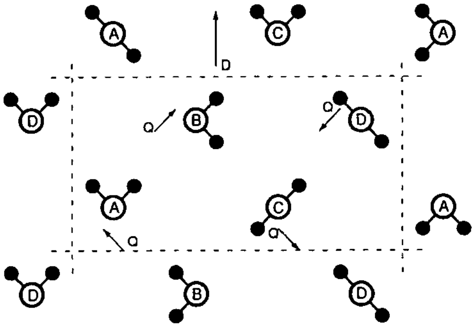

Fig. 3. The icelike two-dimensional six-vertex model. In the F-model the linear molecules have lowest energy. The direct field D points in the vertical (North–South) direction. The staggered field S favors linear molecules aligned along the NE–SW (NW–SE) directions on the A and C (B and D) sublattices, respectively. The quarter field Q points in the NW (NE, SE, SW) direction on the A (B, C, D) sublattices, respectively. The unit cell is shown by dashed lines. For the particular state shown, within the unit cell the energy of the A molecule is $-D-Q$, the energy of the B molecule is $-Q$, and the energies of the C and D molecules are each $-S$.

been a long-term goal of mine to find a dimer model that had both types of transition and this was accomplished by Carlos Yokoi.(19) The bonus was that there is a multicritical point where an O-type transition line crosses a K-type transition line. The temptation to name this an OK multicritical point was just too hard to resist! This OK multicritical point is neither ordinary nor trivial, but is a new kind of critical point that does not fit into the standard multicritical scaling theory.

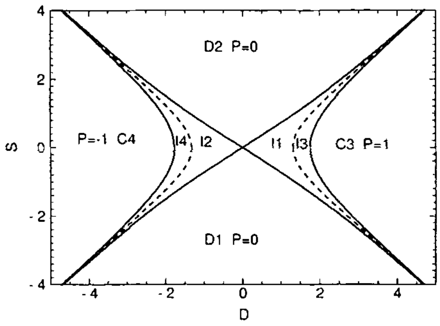

Fig. 4. The F-model phase diagram in D and S fields at $kT = 2\varepsilon/\ln(2)$. Along the $D=0$ line the transition is O-type at $S=0$. When $D \neq 0$ the solid lines show loci of K-type transitions. The regions labeled I (I1–I4) are incommensurate regions with domain walls in which the polarization P varies continuously. The dashed lines show the loci of the new O-type transitions.

Our new dimer model is isomorphic to the six-vertex F-model at the one particular temperature where Baxter showed that the F-model can be solved using the Pfaffian technique.(20) The difference with Baxter's work was that he solved the F-model with two fields, a direct field D and the staggered field S which couples to the order parameter,(21) whereas our recent work(19) solved the F-model in a new quarter field Q as well as the staggered field S, but with no D field. The description of these various fields is shown in Fig. 3.

The phase diagram in the D–S plane obtained by Baxter is shown in Fig. 4, along with a new O-type transition line that will be justified in due course. Our phase diagram(19) in the S–Q plane is shown in Fig. 5a. It was not *a priori* obvious how the two phase diagrams in Figs. 4 and 5a are connected when all three fields (D, S, Q) are varied. This is now shown by the phase diagrams in Figs. 5b–5d.

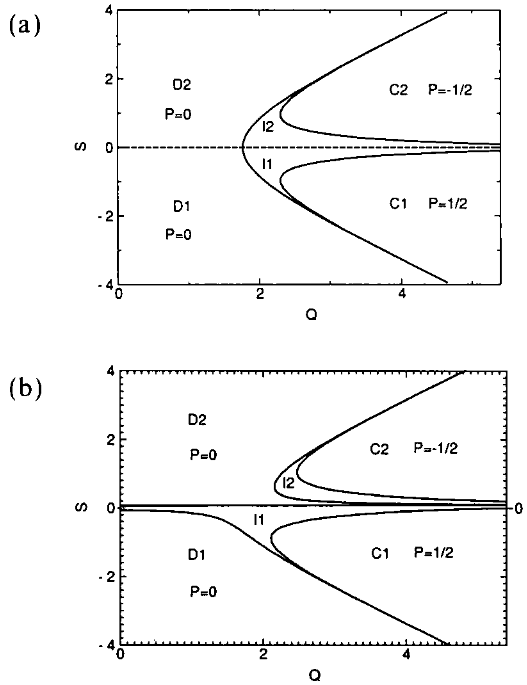

Fig. 5. The F-model phase diagram at $kT = 2\varepsilon/\ln(2)$ in S and Q fields for selected values of the D field. All phase diagrams are symmetric upon replacing Q by $-Q$. The solid lines are loci of K-type transitions. The polarization P varies continuously in the I regions. (a) $D = 0$. The dashed line along the $S = 0$ axis is a locus of O-type transitions. The OK multicritical point is the intersection of the dashed line with one of the solid K-type lines. (b) $D = 0.1$, (c) $D = 1.6$, and (d) $D = 2.0$.

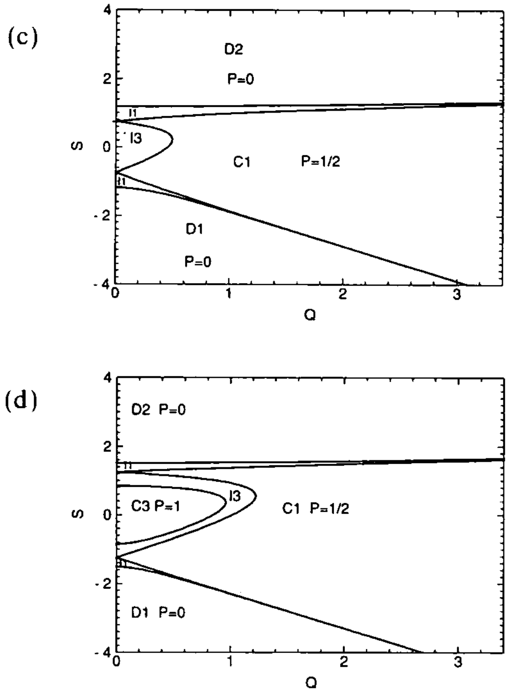

Fig. 5. (*Continued*)

Figure 5b shows the effect of adding a small direct field $D = 0.1$. Compared to the $D = 0$ phase diagram shown in Fig. 5a, continuity between the regions in the two figures is obvious except in the vicinity of the OK multicritical point, which disappears in Fig. 5b. There, the K-type line that goes through the multicritical point tears into two lines, the O-type line in Fig. 5a changes into a K-type line in Fig. 5b and the disordered D2 phase separates the I2 phase from the I1 phase, although this separation becomes very small for large Q. The intersections of the K-type lines with the $Q = 0$ axis on Fig. 5b map into the K-type lines in Fig. 4; this identifies the I1 regions as the same in both figures.

The phase diagrams in Fig. 5 are symmetric upon replacing Q by $-Q$. Also, when D is replaced by $-D$, the phase diagrams in Fig. 5 are obtained by changing S to $-S$ and P to $-P$; this also changes the phases according to C1 ↔ C2, D1 ↔ D2, I1 ↔ I2, and I3 ↔ I4.

As D increases, the I2 and C2 regions are pushed to larger values of Q and the I1 region expands for small S consistent with Fig. 4. The C1 region also moves to smaller Q. It was unexpected, however, that the K-type line that separates the C1 region from the I1 region intersects the

$S=0$ plane. This intersection occurs at one point when $\cosh D=2$ and at two points when $\cosh D>2$, as shown in Fig. 5c. In Fig. 4 these intersections are shown as the dashed lines, which will be identified as O-type lines later.

Figure 4 also shows the perfectly ordered C3 phase with $P=1$ that occurs for $\cosh D>3$. The effect of varying S and Q while holding D fixed at the value $D=2.0$ is shown in Fig. 5d. From the C3 phase, increasing Q takes one through the I3 phase with decreasing P to the partially ordered C1 phase with $P=1/2$.

The phase diagrams in Figs. 4 and 5 were obtained using the well-known Pfaffian method. Details of the calculations are straightforwardly similar to our previous work.(17,19) The evaluation of the central determinant yields

$$\begin{aligned}\det(M_1)=&\,[2\sinh(S)-\sinh(D-i\phi)]^2\\&-4\cosh^2(Q/2)[\cosh(D-i\phi-S)-1]+\sin^2(\theta/2)\end{aligned}\qquad(1)$$

where θ and ϕ are the integration variables when obtaining the free energy from $\log[\det(M_1)]$. The transition lines in the phase diagrams are obtained from the zeros of $\det(M_1)$ when $\phi=0$ and $\theta=0$ or π. Zeros appear for other values of θ in the incommensurate regions indicated by I1–I4 in the figures. No general proof was found that there are no zeros when $\phi\neq 0$, but extensive numerical searching failed to reveal any other zeros. Because D and ϕ only appear in the combination $D-i\phi$, the polarization P can only change in the incommensurate regions.(19)

The new O-type lines that occur in the D–S plane in Fig. 4 when $Q=0$ are characterized by divergences of the quarter susceptibility, which is just the second derivative of the free energy with respect to Q and is therefore the derivative of the quarter polarization P_Q with respect to Q. The behavior of $\det(M_1)$ as these lines are approached is given asymptotically to lowest order as

$$\det(M_1)\sim\theta^2+\phi^2+Q^2\qquad(2)$$

so the quarter susceptibility diverges as $-\ln|Q|$ as Q approaches zero.(17)

How can it be understood that the new O-lines were not previously apparent before a Q field was introduced? At the simplest level, this can be understood by analogy to the O-line at $0=Q=S=D$ when D is swept through $D=0$. In this latter case, it is only the introduction of a staggered field S that yields a staggered susceptibility that diverges at $S=0=D$. The topology of the phase diagram in the vicinity of the new O-line in Fig. 4 is identical to that of the old O-line, with Q replacing S. This topology can be described as each O-type point being the intersection of two K-type lines such that circumnavigating the O-point takes one through a sequence of four phases that alternate as commensurate, incommensurate, commensurate, and incommensurate.

The mathematical development of such O-lines can be understood in terms of the behavior of the zeros of $\det(M_1)$ and the doubling of the unit cell when a new field such as S or Q is imposed. This doubles the angle θ and each band of zeros that occur in the complex $v = \exp(i\phi)$ plane splits into two equal bands. When the system is in the interior of an incommensurate phase such that the real zero occurs when θ is in the center of its band, then the split separates two zeros on the unit circle in the v plane, which gives the asymptotic behavior in Eq. 2.(17,19) Since a variety of fields may be introduced with different-sized unit cells, this discussion suggests that lines within incommensurate regions may contain a countably infinite number of O-type transition points. This property would seem to be consistent with the presence of long-range correlation functions(22) in these phases.

In summary, the new O-lines emphasize that incommensurate regions may be rich in critical points when other fields are considered, but I believe this paper shows that these lines are well understood in physical and mathematical terms. This work also reveals the unusual structure of the phase diagram near the nonstandard OK multicritical point, which still has some mysterious features regarding the correlation functions and scaling behavior.(19) Of course, it is preposterous to suppose that this OK multicritical point would ever be the hole in the dike of current universal theories the way that Onsager's solution of the 2D Ising model was for the classical universal theories. But it is interesting to speculate which kind of theory Onsager would value most today. Although I only knew him from his late 50's, I would be willing to bet that, because of his love and appreciation of variety in science, he would opt for the solutions of particular models rather than universal theory. I feel that he was by temperament somewhat closer to biology than to physics, though his superb mathematical ability cloaked this and made him seem more of a physicist. However, I am sure there are others who disagree with this highly personal and undoubtedly incomplete characterization. Indeed, Lars Onsager was too large to be pigeonholed in any way. Quite likely, when different views of Onsager are compared, the results will be similar to the classic story of three blind men examining an elephant.

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