LA-UR-14-26326

Approved for public release; distribution is unlimited.

Title: Emergent long-range couplings in arrays of fluid cells

Author(s): Abraham, Douglas Bruce

Intended for: Physical Review Letters

Issued: 2014-08-07

Disclaimer:

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By approving this article, the publisher recognizes that the U.S. Government retains nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Emergent long-range couplings in arrays of fluid cells

D. B. Abraham,^{1,2} A. Maciolek,^{3,4,5} and O. Vasilyev^{3,4}

¹*Theoretical Physics, Department of Physics,
University of Oxford, 1 Keble Road,
Oxford OX1 3NP, United Kingdom*

²*Center for Non-linear Studies, Los Alamos National Laboratory, NM 87545, USA*

³*Max-Planck-Institut für Intelligente Systeme,
Heisenbergstr. 3, D-70569 Stuttgart, Germany*

⁴*IV. Institut für Theoretische Physik, Pfaffenwaldring 57,
Universität Stuttgart, D-70569 Stuttgart, Germany*

⁵*Institute of Physical Chemistry, Polish Academy of Sciences,
Kasprzaka 44/52, PL-01-224 Warsaw, Poland*

(Dated: July 14, 2014)

Abstract

We present a system exhibiting extraordinarily long-range cooperative effects, on a length scale far exceeding the bulk correlation length. We give a theoretical explanation of these phenomena based on the mesoscopic picture of phase coexistence in finite systems, which is confirmed by Monte Carlo (MC) simulation studies. Our work demonstrates that such action-at-a-distance can occur in classical systems involving simple or complex fluids, such as colloid-polymer mixtures, or ferromagnets.

PACS numbers: 05.40.-a, 05.50.+q, 68.05.Cf, 68.35.Rh

Can correlation effects in a fluid confined in big but finite compartments linked by small openings, such as shallow channels, occur over distances much larger than the bulk correlation length? Recently, Gasparini and co-workers [1, 2] have demonstrated such rather striking “action-at-a-distance” effects in a two-dimensional array of microscopic boxes filled with superfluid ^4He and linked by either channels or a uniform film. The measurements of several responses show that under certain conditions these boxes can be strongly coupled. What seems to be crucial in this work is the size of boxes and connectors and the vicinity of the critical point [2]. Perron et al [2] suggested that observed action-at-a-distance effects might be a more general feature of systems with phase transitions than is usually supposed, a view which we confirm in this work for uniaxial classical ferromagnets and their analogs (simple fluids or binary mixtures in the lattice gas approximation, all belonging to the Ising model universality class of critical phenomena).

In the lattice gas picture the space occupied by the system is divided into boxes, either vacant or containing a single molecule, unit maximal occupation is achieved by a judicious choice of the box size. The state of a box at position i with integer coordinates is labeled by a spin variable $\sigma_i = \pm 1$. In the absence of bulk ordering field, a configuration $\{\sigma\}$ of such (classical) spins has an energy

$$E(\{\sigma\}) = -J \sum_{\langle ij \rangle} \sigma_i \sigma_j. \quad (1)$$

The sum $\langle ij \rangle$ is taken over all nearest-neighbor pairs ij of spins and J is the coupling constant. Since the spins are assumed to be in thermal equilibrium with a bath at temperature T , the probability of any spin configuration $\{\sigma\}$ is given by $p(\{\sigma\}) = Z^{-1} \exp(-\beta E(\{\sigma\}))$, where $\beta = 1/k_B T$ (k_B is the Boltzmann constant), and Z is the normalization. The Helmholtz free energy from the formula $F = -(1/\beta) \ln Z$. For dimensionality $d \geq 2$, it is known that such systems undergo phase transition to a low-temperature, magnetically ordered (dense) state [3]. For the square lattice, the critical value of $K = J\beta$ is given by $K_c = (1/2) \ln(1 + \sqrt{2}) \approx 0.440687$ [3]. When $d = 3$, various estimations are available [4]; $K_c(d = 3) \approx 0.2216544(3) \sim K_c(d = 2)/2$. Thus a $3d$ lattice orders more easily (higher critical temperature); this is compatible with Griffiths’ correlation inequalities [5].

Although an infinite system size is mandatory for a sharp transition [3], Fisher and Privman [6] gave a simple physical picture, denoted FP, of great elegance, which captures the effect of finite size in a predictive way. Let us see how it works for a strip geometry in

FIG. 1. (Color online) Side view of an Ising system comprised of two cubic lattice boxes of a side L_0 connected by a $L \times M$, $L \gg M$ strip. We assume $L_0 \gg M$.

$d = 2$ below the bulk critical temperature T_c . If we sum out fluctuations up to a certain length scale, a process often called coarse-graining and take as a scale the bulk correlation length, then a typical configuration in a strip is one with regions of alternating (+) and (−) magnetization, with a magnitude roughly the spontaneous magnetization m , separated by domain walls (Fig. 1). The statistical weight of such a domain wall is taken to be $\tilde{w} = \exp(-M\tau)$, where M is the width of the strip and τ is the reduced (by a factor of $k_B T$) interfacial tension or a domain wall free energy. The configurational entropy is estimated by treating the domain walls as point particles on the line which are strictly avoiding. Using this simple picture, we can calculate exactly the pair correlation function $G(x)$ of two spins in the same edge of a strip separated by a distance x (we use throughout a lattice constant as a length unit). The spins are taken to be parallel (anti-parallel) if there is an even (odd) number of domain walls separating them which gives [7]:

$$G(x) \propto \left(\frac{1 - \tilde{w}}{1 + \tilde{w}} \right)^x. \quad (2)$$

Equation (2) should be complemented with the prefactor $m_e(M)$, which becomes the edge magnetization as $M \rightarrow \infty$ [8]. Its value is a manifestation of fluctuations on the scale of the bulk correlation length ξ_b and thus is not accessible in FP. For sufficiently wide strips and fixed temperature below $T_c(d = 2)$ such that $M\tau \gg 1$ ($\tilde{w} \ll 1$), Eq. (2) can be simplified to give a pure exponential decay:

$$\begin{aligned} G(x) &= m_e^2(M) \exp \left[-x \log \left(\frac{1 + \tilde{w}}{1 - \tilde{w}} \right) \right] \\ &\sim m_e^2 \exp \left[-2x\tilde{w}(1 + O(\tilde{w}^2)) \right]. \end{aligned} \quad (3)$$

It is noteworthy that in the FP picture the decay length of correlation function $G(x)$ *diverges exponentially* with M . We now compare this prediction with the result of exact calculation

for the full Ising strip [9]. We find agreement in the asymptotic behavior of $G(x)$ provided $\tilde{w}$ in eq. (3) is replaced by $w = (\sinh 2K)^{-1} \sinh(\tau) e^{-M\tau}$. This is easy to understand if we note that the simple Helmholtz fluctuation estimate expressed by $\tilde{w}$ must be modified to include the point tension (a $2d$ analogue of line tension) and this we calculate exactly in confirmation. In the exact result of Ref. [9], there is an additional contribution to $G(x)$ due to fluctuations on the scale of the bulk correlation length ξ_b . Because in $2d$ Ising model $\xi_b = 1/\tau$, this contribution is relatively negligible provided $w \ll \tau$. This gives us a criterion for the validity of our theory which confirms naïve expectations. Stated another way, the recapture of long-range order is achieved *not* through bulk correlation length related phenomena, but rather by the emergence of a new length scale which diverges exponentially fast as $M \rightarrow \infty$, as $\exp(M\tau)$. We now apply the FP idea with improved statistical weight w

FIG. 2. (Color online) (a) Geometry of two-dimensional array of $N_0 \times N_0$ cubes of size L_0 connected by strips (channels) of length L and thickness M . (b) The states of boxes are described by the spin variable $S_j = \pm 1$; the nearest-neighbor boxes interact with the interaction energy $K_1 S_i S_j$.

of the domain wall, which we term *enhanced* Fisher-Privman theory (EFP), to the scheme of Fig. 1: the pair of cubic lattice boxes of side L_0 is coupled by an Ising strip of dimension $L \times M$, with $L_0 \gg M$. For $T < T_c(d = 2)$, the picture which emerges is one with a sequence of domain walls crossing the strip, but none inside the boxes, because they would be of the size $L_0 \times L_0$ and controlled by a higher surface tension, thus negligible. Because $K_c(d = 3) \sim K_c(d = 2)/2$ and boxes are large, we expect that the state of each box is either magnetized up or down and assignment can be described on this level of coarse graining by a variable $S_j = \pm 1$ for each box, $j = 1, 2$. The Boltzmann factor for a given assignment in

place of argument of the S_j is thus [7]

$$Z = Z_e^{(1+S_1 S_2)/2} Z_o^{(1-S_1 S_2)/2} = A e^{K_1 S_1 S_2} \quad (4)$$

where Z_o (resp. Z_e) is the partition function for an odd (resp. even) number of domain walls and the interaction constant K_1 is given by

$$e^{2K_1} = \frac{Z_e}{Z_o} = \frac{1+t^L}{1-t^L}, \quad t = \frac{1-w}{1+w}. \quad (5)$$

It is crucial to note that the temperature does not enter in the usual Boltzmann way in K_1 . We can assemble such bonds, assumed mutually independent, and boxes to make up a

FIG. 3. (Color online) The phase diagram of the two-dimensional “network” lattice shown in Fig. 2. (a) The critical value of the length of connecting strips L_c depends on the surface tension τ and the width M of the strip. Network is ordered in the region which lies below the critical surface $L_c(\tau, M)$. (b) The phase diagram in the scaling limit $M, L \rightarrow \infty$ and $\tau \sim T - T_c(d = 2) \rightarrow 0$; $a = 2^{-1} \ln(1 + \sqrt{2})$.

“network” lattice. For an analogous set up to that of Gasparini and co-workers we take a two dimensional array as illustrated in Fig. 2. The intriguing possibility is that K_1 could satisfy $K_1 > K_c(d = 2)$ by, e.g., adjusting the width or length of the strip at a fixed temperature $T < T_c(d = 2)$. Solving above with $K_1 = K_c(d = 2)$ gives a critical value L_c where $L_c \ln(1/t(M, \tau)) = \ln(1 + \sqrt{2})$. With $L < L_c(M, \tau)$, the system is subcritical and hence *ordered*. Because w is small, this implies that $2L_c w = \ln(1 + \sqrt{2})$; thus L_c diverges as $\exp(M\tau)$. In the EFP (contrary to FP), this result scales near the $2d$ critical point. Using scaling variables $L_c \tau$ and $M\tau$ we obtain for the network critical point $L_c \tau e^{-M\tau} = 2^{-1} \ln(1 + \sqrt{2})$. Although it is not well visible on the scale of Fig. 3(a), the critical surface

FIG. 4. (Color online) MC simulation data for a $2d$ array of 10×10 squares with the side $L_0 = 100$ connected by strips of the length $L = 100$ and the width $M = 10$: (a) the spin-spin correlation function $G(x) = \langle \sigma(0)\sigma(x) \rangle$ as a function of the distance x along the center of the channel (red solid line in Fig 2) for various couplings $K = 0.46, 0.53, 0.55, 0.56, 0.58$ (results for smaller K are not accessed from the EFP theory), (b) the susceptibility χ and (c) the specific heat C of the system as a function of K . The specific heat for a single square (dashed line) is given for comparison. The values of χ at temperatures for which the correlation functions are shown in (a) are highlighted by points with the same color as the corresponding curves $G(x)$.

$L_c(M, \tau)$ displays an interesting feature: for sufficiently wide strips, the critical value L_c of strip length is a non-monotonic function of temperature. This offers a possibility to tune the collective behavior of boxes by varying T . If the size of connecting strips is suitable chosen, the initially correlated boxes become uncorrelated upon increasing T but then correlated again sufficiently close to $T_c(d=2)$.

We have tested the application of EFP theory by MC simulation. The averaging has been performed over 10^5 of MC steps. Each hybrid MC step consists of a flip of Wolff

FIG. 5. (Color online) Magnetic susceptibility of the two-dimensional array of $N_0 \times N_0$ cubes of size L_0^3 , connected by channels of size $L \times W \times W$ as function of K for $N_0 = 10$, $L_0 = 20$, $W = 4$ and various $L = 6, 10, 20, 30, 40$.

cluster and application of Metropolis updates to a randomly chosen quarter of all spins in the system [10]. For a $1d$ array of $2d$ Ising boxes and strips we observe (not shown here) that the pair correlation function below $T_c(d=2)$ has plateaux, which confirms our expectation that boxes are ordered. As predicted, these plateaux values approach zero following the Ising correlation function law $G(x) \simeq m^2(\tanh K_1)^{|x/L|}$ with $\tanh K_1 = t^L$ where m is the (spontaneous) magnetization in the box. We have also performed MC simulation of a $2d$ array of squares. We find that, below certain temperature, the pair correlation function $G(x)$ along the line connecting centers of squares via the channels (red line in Fig. 2) does not decay to zero. This is a clear manifestation of the existence of order in the network. For example, the system for which the MC data are shown in Fig 4, undergoes the (rounded in the finite system) ordering transition at $K \approx 0.55$, which agrees perfectly with the prediction from the EFP theory. This transition lies *below* the critical point of the $2d$ Ising model, as implied by Griffiths inequalities; this is because such a lattice has been perforated to arrive at the network model. It will be accompanied by a divergent susceptibility; in the MC simulation data shown in Fig 4(b) one can see a peak located at $K_m \simeq 0.55$. The ghost of the rounded phase transition in the connecting strips appears as a shoulder (red dot in Fig. 4(b)). No signal of network ordering is seen in the specific heat. Our EFP arguments also apply to boxes connected by rods. But in this case there are no exact results available for the quantity w . We must resort to simulation for its evaluation, as will be detailed elsewhere. The long-range coupling between boxes should occur, much as in the results

reported above. To confirm this anticipation, we have performed MC simulation of the two-dimensional array of $N_0 \times N_0$ cubes of size L_0^3 connected by channels of the length L and cross-section $W \times W$ (the top view of this system is shown in Fig. 2(a)). The magnetic susceptibility χ of such a system as function of K displays features similar to these of the $2d$ array of squares connected by strips. We observe, as expected, that the only effect of the system size N_0 on χ is to increase the height of the peak at the ordering transition point. The cube size L_0 affects the shoulder of the curve which is the remnant of the $3d$ bulk transition, but does not change the location of the peak. The location of the ordering transition point is determined entirely by the channel geometry - for dependence on L see Fig. 5.

We have introduced in this work a theory supporting the intriguing suggestion by Perron et al [1] that the action at a distance effect which they observed experimentally in superfluid ^4He might be a *widespread* consequence of phase transitions and critical phenomena. Our theory, which applies to classical lattice gases and their analogues, has a key ingredient: the extended Fisher-Privman theory of finite size effects in first order phase transitions [6]. In the network Ising model constructed from the $2d$ array of boxes and connecting strips, we see that the parameters can be tuned to produce long-range order, in itself not perhaps surprising, but with *extraordinarily long connecting links*; this diverges exponentially with strip width, on a scale of the inverse surface tension. Thus, ordering between boxes is feasible over length scales of many thousands of molecular diameters, which maybe tested in experiments. The action-at-a-distance which we have described depends on the boundary conditions imposed on the connecting strips. For instance, for fixed spins, say all up, there are no long-range cooperative effects whereas, for wetting boundary conditions in the partial wetting regime there are indeed long range effects; this opens an intriguing possibility of tunability and could be used as a microfabricated device in soft matter and biophysics experiments [11] Systems with strips joined through their edges, which also display long-range order [12, 13], generate markedly different physics from those we considered where the strips are joined through their extrema at nodes (see Fig. 2).

DBA acknowledges with gratitude the support of Prof. S. Dietrich at the MPI Stuttgart, where parts of this work were done.

-
- [1] J. K. Perron, M. O. Kimball, K. P. Mooney, and F. M. Gasparini, *Nature Physics* **6**, 499 (2010).
- [2] J. K. Perron and F. M. Gasparini, *Phys. Rev. Lett.* **109**, 035302 (2012).
- [3] L. Onsager, *Phys. Rev.* **65**, 117, (1944); R. Peierls, *Mathematical Proceedings of the Cambridge Philosophical Society* **32**, 477 (1936).
- [4] C. Ruge, P. Zhu, and F. Wagner *Physica A* **209**, 431 (1994).
- [5] R. B. Griffiths, *J. Math. Phys.* **8**, 478 (1967).
- [6] V. Privman and M. E. Fisher, *J. Stat. Phys.* **33** 385, (1983).
- [7] See Supplemental Material at [] for derivation of Eqs. (2) and (5).
- [8] B. M. McCoy and T. T. Wu, *Phys. Rev.* **162**, 436 (1967).
- [9] D. B. Abraham, *Stud. Appl. Math.* **50**, 71 (1971).
- [10] D. P. Landau and K. Binder, *A Guide to Monte Carlo Simulations in Statistical Physics* (Cambridge University Press, London, 2005), p. 155.
- [11] D. Bartolo and D. G. A. L. Aarts, *Soft Matter*, **8**, 10530, (2012), and references therein.
- [12] M. E. Fisher, *Nature Physics* **6**, 483 (2010).
- [13] H. Au-Yang and M. E. Fisher, *Phys. Rev. E* **88**, 032147 (2013).