Scaling in Modulated Systems and Re-entrance of Order

O. Portmann, A. Vindigni, and D. Pescia
Laboratorium für Festkörperphysik, ETH Zürich, 8093 Zürich, Switzerland
(Dated: July 22, 2009)

We propose scaling invariance of \(d\)-dimensional modulated systems, in which modulation of order is produced by a frustrating long-ranged interaction decaying spatially with some power \(\alpha\) of the inverse distance. Over the entire temperature range in which a modulation length is defined, measurable physical quantities acquire scaling exponents. One outcome of the scaling analysis is the existence, for \(\alpha > d\), of an anomalous state in which the tendency of matter to order increases with temperature. We suggest that this anomaly is responsible for a re-entrance phenomenon in two-dimensional dipolar frustrated magnetic systems.

Modulation of order is a general motive in chemistry, biology, and physics [1, 2]. The modulated order parameter may represent quantities as diverse as the spin density \([3, 4, 5]\), the charge density in any type of strongly correlated classical or quantum system \([2, 6, 7]\), the volume fraction of diblock copolymers, the concentration of amphiphilic molecules and other chemical species \([2, 3]\), or dipolar bosons in an optical lattice \([10]\). However, modulated systems tend to show common characteristics such as the morphology of the various patterns and the occurrence of transitions among them \([1]\), as in liquid crystals \([11]\) or two-dimensional melting phenomena \([12]\). This tendency to common behavior indicates universal underlying principles \([2, 3, 8, 12]\).

In this Letter, we consider \(d\)-dimensional systems where the spatial modulation of the order parameter is produced by the competition of a short-ranged interaction favoring local order and a weak but long-ranged frustrating interaction decaying with some power \(\alpha\) of the inverse distance. We propose that these systems are scaling invariant along the entire temperature range where some characteristic modulation length \(L\) exists. Accordingly, measurable physical quantities are characterized by scaling exponents which only depend on fundamental symmetries of the system, like \(d\) and \(\alpha\). One outcome of the scaling analysis is that modulation of order is only stable if \(\alpha\) is less than an upper limit dependent on \(d\). A further outcome is that, for \(\alpha > d\), the tendency of the modulated system to order – measured by the compression modulus \(B\) – can increase with temperature. Among the systems that are predicted to show this anomaly are a model of two-dimensional Coulomb frustrated phase separation \([8]\) and two-dimensional ferromagnets frustrated by the dipolar interaction \([3, 4, 5]\). We report experimental data on perpendicularly magnetized ultrathin Fe films on Cu(100) (\(\alpha = 3 > d = 2\)) \([14, 15]\) that confirm the anomalous growth of \(B\) with temperature. This growth of \(B\) goes along with a re-entrance \([16]\) of the more ordered stripe pattern \([14]\).

Following Muratov \([2]\), a universal effective Landau-Ginzburg-Wilson (LGW) Hamiltonian (per volume) \(\mathcal{H}\) describing systems frustrated by a long-ranged interaction reads \([17]\)

\[
\mathcal{H}(\phi(x), T) = \frac{1}{V_D} \int \left\{ \frac{1}{2} \phi_0^2 (\nabla \phi(x))^2 - T \phi_0^4 \left[ \frac{\phi^2(x)}{6} - \frac{\phi^4(x)}{12} \right] \right\} d^Dx \\
+ \frac{1}{V_D} \lambda \phi_0^2 \int \phi(x) \phi(x') G_\lambda(|x-x'|) d^Dx d^Dx'
\]

(1)

where \(x\) being a vector in a \(D\)-dimensional space and \(V_D\) the volume of the system. Without the long-ranged interaction (\(\lambda = 0\)), this functional has macroscopic phase separation, with an order parameter \(\phi_0\) appearing below a critical temperature \(T_C\). The field \(\phi(x)\) is a scalar order parameter (rescaled with \(\phi_0\)). The gradient term in the first line mimics the short-ranged exchange contribution. It is inaccurate for sharp walls at low temperatures, where a more suitable expression exists \([4]\). For large separations \(|x-x'|\), the kernel of the long-ranged interaction \(G_\lambda(|x-x'|)\) is assumed to behave asymptotically like \(|x-x'|^{-\alpha}\). \(\alpha = 1\) corresponds to the Coulomb interaction and gives rise to the Coulomb Frustrated Ising Ferromagnet (CFIF) \([6]\). The kernel of the Dipolar Frustrated Ising Ferromagnet (DFIF) \([20]\) has a more complicated structure but can also be treated with Eq. \([17]\). We assume \(\frac{\lambda}{\phi_0^2} \gg 1\) and hence a mono-dimensional modulation of length \(L \gg 1\) along a coordinate \(x_1\) in a system with a large extent in \(d\) dimensions. In the remaining \(D - d\) dimensions, the system is assumed to have a finite thickness \(O(\delta) \ll L\).

At a given temperature \(T\), we expect that a minor change of the modulation length \(L\) will have a limited effect on the order-parameter profile \(\phi(x)\). We therefore postulate the invariance of \(\phi(x)\) with respect to a scaling of all lengths by a constant \(u\):

\[
\phi(u \cdot x_1, u \cdot L) = \phi(x_1, L)
\]

(2)

i.e. \(\phi\) to be a function of one single variable \(\tilde{x}_1 = \frac{x_1}{L}\); we thus introduce \(\tilde{\phi}(\frac{\tilde{x}_1}{L}) \approx \phi(x_1, L)\). Considering only the asymptotic behavior of \(G_\lambda\), we can now transfer the \(L\)-dependence of \(\mathcal{H}\) to prefactors \(L^\Delta\) with suitable scaling exponents \(\Delta\). Per unit volume, the part due to the exchange interaction scales like \(O(\phi_0^2 L^\Delta)\) and the part due to the long-ranged interaction like \(O(\lambda \phi_0^2 L^\Delta)\), with \(\Delta_f = -1\) (sharp walls between adjacent domains) or \(-2\)
the solution of the equation \(\partial\phi/\partial L\) for the \((D = 3, d = 2)\)-DFIF. For \(\Delta_j = -1\) \((T\approx0)\), we have \(\Delta_{L_j} = \infty\), which suggests an exponential dependence on \(J/\lambda\) (confirmed by detailed computations 18). At higher temperatures, \(L_j\) only depends linearly on \(J/\lambda\). For \(J/\lambda \gg 1\), this means a decrease of the modulation length by several orders of magnitude 20. The compression modulus \(B_j(T)\) 14, 21 also called Young modulus 22 measures the energy cost associated with deviations from the equilibrium modulation length \(L_j(T)\) and is given by

\[
B_j(T) \equiv L_j^2 \cdot \frac{\partial^2 \Phi(L, T)}{\partial L^2} \Bigg|_{L_j(T)},
\]

which, in virtue of the Hellmann-Feynman theorem, is equivalent to

\[
L_j^2 \left( \frac{\partial^2}{\partial L^2} \Phi(\phi(x), L, T) - \frac{1}{T} \left( \frac{\partial}{\partial L} \Phi(\phi(x), L, T) \right)^2 \right) \bigg|_{\phi(x) = L_j}.
\]

Positivity of \(B_j\) is a necessary condition for the stability of the modulated state. The scaling analysis, applied to Eq. 5, shows that \( \langle \frac{\partial^2}{\partial L^2} \Phi(\phi(x), L, T) \rangle \sim \lambda(J/L) \frac{2-\alpha_\nu}{2-\alpha_{\nu}}\langle \frac{\partial^2}{\partial L^2} \Phi(\phi(x), L, T) \rangle \bigg|_{\phi(x) = L_j} \), so that positivity of \(B_j\) in the limit \(J/\lambda \gg 1\) requires

\[
\alpha - d < -\Delta_j.
\]

This result generalizes the one obtained in Ref. 13 for \(T = 0\) and \(d = 1\). When the modulated state is stable, \(B_j \approx \lambda \cdot \phi_0^2 \cdot L_j^{\Delta B_j}\) (for \(\Delta B_j\), see fifth column). Note that

\[
\begin{array}{c|c|c|c|c}
(D, d) & \Delta_{L_j}^{-1} & \Delta_{L_j}^{0} & \Delta_{B_j}^{-1, -2} \\
\hline
\text{DFIF}(3, 3) & \frac{1}{d-\alpha+1} & \frac{1}{d-\alpha+2} & d-\alpha \\
\text{DFIF}(3, 2) & 3 & \infty & 1 & -1 \\
\text{CFIF}(3, 3) & 1 & \frac{1}{d-\alpha+1} & 2 & 1 \\
\text{CFIF}(3, 2) & 1 & \frac{1}{d-\alpha+1} & 1 & 1 \\
\end{array}
\]

TABLE I: Scaling exponents \(\Delta_{L_j}\) and \(\Delta_{B_j}\) for general \(d\) and \(\alpha\) and for some special cases. The asymptotic long-range interaction of the \((D = 3, d = 3)\)-DFIF does not follow a simple \(|x - x'|^{-\alpha}\)-law. It therefore cannot be assigned a value of \(\alpha\) and the exponents have to be determined separately.
excitations are responsible e.g. for the Landau-Peierls instability that prevents order in $D = 3$ for $d = 1, 2$ \cite{2, 3, 4}. Averaging the perturbed functional $\mathcal{H}[\phi(x_1 - \epsilon_{hk} \cos(kx_2) \cdot \cos(hx_1)), L, T]$ expanded up to order $\langle \epsilon_{hk} \rangle^2$ and powers $(h \cdot k)^n$ with $n \leq 4$ using the equipartition theorem $\langle \epsilon_{hk} \rangle^2 \approx \frac{T}{\Omega^2} \frac{1}{\Omega^2 + \lambda\hbar^2 - \omega^2}$ \cite{4} and summing over $(h, k) \leq 1/L$, we recover an effective mono-dimensional functional in which the exchange energy and the energy of the long-ranged interaction have, to leading order, the same scaling behavior, albeit with modified coupling constants $J \to J + O(T)$ and $\lambda \to \lambda + O(T)$. This shows that scaling is conserved at finite temperatures and that the scaling dimensions are not changed by small perturbations. In particular, the Landau-Peierls instability destroys long-range positional stripe order but the loss of order proceeds in such a way that the modulation length and the compression modulus $B_3$ remain well-defined quantities also at finite temperatures \cite{22}.

Third, the compression modulus $B_\Omega$ of perpendicularly magnetized ultrathin Fe films on Cu(100) \cite{14} shows the anomalous enhancement with temperature predicted by scaling. In these systems, a strong magnetic anisotropy aligns the spins perpendicular to the film plane. Accordingly, the exchange interaction is frustrated by the (isotropic part) of the long-range dipolar interaction, i.e. it is a $(d = 2)$-DFIF and has a characteristic modulation length \cite{1, 14}, up to a temperature $T_C$ at which $\phi_0$ vanishes. The data points in Fig. 2 display $B_\Omega(T)$ as determined experimentally using the scaling result. The solid curve is obtained from fits to $\phi_0(T)$ and $L_\Omega(T)$. The decrease of $\phi_0(T)$ and the appearance of mobile stripes just below $T_C$ makes the experimental determination of $B_\Omega(T)$ difficult. Note that in zero applied magnetic field, the low-temperature state is striped, but transforms upon temperature increase into a more disordered labyrinthine pattern, displayed left in Fig. 2 \cite{1, 13}. This transformation has also been reported in a micrometer-thick film, a $(d = 3)$-DFIF \cite{1}. What is new in the ultrathin limit is that the labyrinthine pattern transforms back into the more ordered stripe pattern (pictured right in Fig.2) when the temperature is increased. This re-entrance of order proceeds in the temperature region (light shaded area) where the widespread local configurations in the labyrinthine pattern with deviations from the equilibrium stripe width are increasingly

![Figure 1](image1.png)

**FIG. 1:** (a): (color online). Profile of the order parameter in a $(d = 2)$-DFIF ($\delta = 1$, $\Omega/J = 0.08$) for different values of $L$ ($0.7L_\Omega$, $L_\Omega$ and $1.3L_\Omega$). (b): Collapse of profiles onto one single curve in plot versus $x$. (c): Numerical $B_\Omega(T)$ for the $(d = 2)$-DFIF (black curve, $\delta = 1$, $\Omega/J = 0.08$) together with the scaling result (light line). (d): Numerical and scaling curves $B_\Omega(T)$ for the $(d = 3)$-DFIF ($\delta = 200$, $\Omega/J = 0.08$) and the $(d = 2, 3)$-CFIF (inset, $\delta = 1.200$, $Q/J = 0.0001$). Note that at low temperatures the wall is very thin and the numerical results are unstable. Light circles on the vertical axis indicate the values of $B_\lambda$ at $T = 0$ obtained analytically.

![Figure 2](image2.png)

**FIG. 2:** Experimental $B_\Omega(T)$ for an ultrathin Fe film on Cu(001) obtained by dividing $\phi_0(T)$ by $L_\Omega(T)$. The solid line results from fits to $\phi_0(T)$ and $L_\Omega(T)$ respectively and is a guide to the eye. The temperature range with stripe domains is shaded, the dark shade indicating mobile stripes. The two insets show SEMPA (Scanning Electron Microscopy with Polarization Analysis) images of the labyrinthine and the stripe pattern. The length of the white bar is 10 $\mu$m, the thickness of the film 1.95 ML.
penalized by the increasing $B_\Omega(T)$. This suggests that the unusual re-entrance of stripe order is a response of the system to the extra elastic energy introduced by the increase of $B_\Omega(T)$. Note that the experimental temperature dependence of the stripe width is smooth in the temperature range where the labyrinth-to-stripe transformation proceeds, which confirms the predicted insensitivity of $L_\Omega$ to deviations from mono-dimensional order. In the vicinity of $T_C$ (dark shaded area), the stripes become mobile. This mobility coincides with the range of temperatures where $B_\Omega(T)$ is expected to decrease and, accordingly, can no longer counteract thermal excitations which destroy stripe order.

In conclusion, we have presented a scaling procedure that is generally applicable to any pattern-forming system and allows re-writing the Hamiltonian as a functional of a scaling-invariant scalar field and rescaled coupling constants $J \rightarrow JL^\Delta$, and $\lambda \rightarrow \lambda L^\Delta$. The temperature dependence of $L_\lambda$, which may vary by several orders of magnitude, is thus expected to propagate in a non-trivial way to any thermodynamic observable. Among other possible implications, scaling predicts a yet undetected state of matter with a compression modulus increasing with temperature. This anomaly provides a simple explanation for an anomalous sequence of patterns observed in ultrathin ferromagnetic films. We show that this explanation proceeds, which confirms the predicted insensitivity of $L_\Omega$ to deviations from mono-dimensional order. In the vicinity of $T_C$ (dark shaded area), the stripes become mobile. This mobility coincides with the range of temperatures where $B_\Omega(T)$ is expected to decrease and, accordingly, can no longer counteract thermal excitations which destroy stripe order.

We acknowledge the financial support of ETH Zurich and the Swiss National Science Foundation as well as fruitful discussions with P. Politi.

[1] M. Seul and D. Andelman, Science 267, 476 (1995) and references therein.
[2] C. B. Muratov, Phys. Rev. E 66, 066108 (2002) and references therein.
[3] T. Garel and S. Doniach, Phys. Rev. B 26, 325 (1982).
[4] A. B. Kushuba and V. L. Pokrovsky, Phys. Rev. B 48, 10335 (1993); Ar. Abanov, V. Kalatsky, V. L. Pokrovsky, and W. M. Saslow, Phys. Rev. B 51, 1023 (1995).
[5] Kwok-On Ng and D. Vanderbilt, Phys. Rev. B 52, 2177 (1995); A. D. Stoycheva and S. J. Singer, Phys. Rev. Lett. 84, 4657 (2000); K. De'Bell, A. B. MacIsaac, and J. P. Whitehead, Rev. Mod. Phys. 72, 225 (2000); D. G. Barci and D. A. Stariolo, Phys. Rev. Lett. 98, 206004 (2007); S. A. Pighin and S. A. Cannas, Phys. Rev. B 75, 224433 (2007) and references therein.
[6] M. Grousson, G. Tarjus, and P. Viot, Phys. Rev. E 62, 7881 (2000); M. Grousson, V. Krakoviack, G. Tarjus, and P. Viot, Phys. Rev. E 66, 026126 (2002).
[7] Z. Nussinov, J. Rudnick, S. A. Kivelson, and L. N. Chayes, Phys. Rev. Lett. 83, 472 (1999) and Z. Nussinov, [ArXiv:cond-mat/0505654].
[8] R. Jaime, S. Kivelson, and B. Spivak, Phys. Rev. Lett. 94, 056805 (2005); C. Ortix, J. Lorenzana, and C. Di Castro, Phys. Rev. Lett. 100, 246402 (2008).
[9] H. M. McConnell, Annu. Rev. Phys. Chem. 42, 171 (1991).
[10] C. Menotti, C. Trefeger, and M. Lewenstein, Phys. Rev. Lett. 98, 235301 (2007).
[11] C. Harrison et al., Phys. Rev. E 66, 011706 (2002) and references therein.
[12] K. J. Strandburg, Rev. Mod. Phys. 60, 161 (1988) and references therein.
[13] A. Giuliani, J. L. Lebowitz, and E. H. Lieb, Phys. Rev. B 74, 064420 (2006); M. Biskup, L. Chayes, and S. A. Kivelson, Comm. Math. Phys. 274, 217 (2007).
[14] O. Portmann, A. Vaterlaus, and D. Pecia, Nature 422, 701 (2003).
[15] O. Portmann, A. Vaterlaus, and D. Pecia, Phys. Rev. Lett. 96, 047212 (2006).
[16] T. Narayanan and A. Kumar, Physics Reports 249, 135 (1994); L. Greer, Nature 404, 134 (2000); N. Schupper and N. M. Shnerb, Phys. Rev. E 72, 046107 (2005).
[17] The coupling constant $\lambda$ amounts to $\Omega \equiv \mu_0 (2g\mu_B S)^2 \cdot (4\pi a^3)^{-1}$ in the dipolar case and to $Q \equiv g^2 \cdot (4\pi e_0)^{-1}$ for the Coulomb interaction respectively. All lengths in Eq. 1 are in units of the lattice constant $a$. For convenience, we set $a_B = 1$.
[18] L. D. Landau and E. M. Lifshitz, Statistical Physics, Pergamon Press, 1980, p. 49.
[19] E. Nielsen, R. N. Bhatt, and D. A. Huse, Phys. Rev. B 77, 054432 (2008).
[20] A. Vindigni, N. Saratza, O. Portmann, D. Pecia, and P. Politi, Phys. Rev. B 77, 092414 (2008).
[21] D. Sonnette, J. Physique 48, 151 (1987).
[22] M. Klemán and O. D. Lavrentovich, Soft Matter Physics, Springer-Verlag, 2003, p. 144.
[23] In order to obtain more precise MFA results at lower temperatures, we use the Bragg-Williams approximation for the entropy (cf. Ref. [22]) in Eq. 1.
[24] R. de Koker, W. Jiang, and H. M. McConnell, J. Phys. Chem. 99, 6251 (1995).
[25] F. Cinti, O. Portmann, D. Pecia, and A. Vindigni, Phys. Rev B 79, 214434 (2009).