Two universality classes for the displacive phase transitions in perovskites

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Perovskites like LaAlO₃, (or SrTiO₃) undergo displacive structural phase transitions from a cubic crystal to a trigonal (or tetragonal) structure. For many years, the critical exponents in both these types of transitions have been fitted to those of the isotropic three-components Heisenberg model. Recent field theoretical accurate calculations showed that this is wrong: the isotropic fixed point of the renormalization group (RG) is unstable, and RG iterations flow either to a ‘cubic’ fixed point or to a fluctuation-driven first-order transition. These distinct flows correspond to two distinct universality classes, identified by the symmetry of the ordered structures below the transitions. Here we show that perovskites which become trigonal or tetragonal belong to these two universality classes, respectively. The close vicinity of the isotropic and cubic fixed points explains the apparent wrong observations of a single universality class, but also implies the existence of slowly varying effective material-dependent exponents. For the tetragonal case, these effective exponents can have the ‘isotropic’ values before crossing to the first-order transition. We propose dedicated experiments to test these predictions. We also expect a similar splitting of the universality classes in any situation in which two (or more) fixed points compete for stability.

Introduction.– A continuous (second-order) phase transition is accompanied by strong fluctuations of its order parameter, with a correlation length ξ which diverges as the temperature T approaches its (non-zero) critical value $T_c$, $\xi = \xi_0 |t|^{-\nu}$, where $t = (T/T_c - 1)$, $\xi_0$ is of order 1 and ν is the ‘critical exponent’ of the correlation length. (Here and below lengths, including $\xi$, are measured in units of the lattice constant.) The divergence of $\xi$ implies that at $T_c$ the system is correlated on all length scales. A similar multitude of length scales occurs, e.g., at fully developed turbulence, quantum field-theory, the Kondo effect, and the self-avoiding-walks for polymers. Below $T_c$, the $n$–component order parameter itself (e.g., the magnetization of a ferromagnet, the staggered polarization of an antiferroelectric crystal, or the vector Q along the rotation axis of the oxygen octahedra in the perovskites, with a magnitude proportional to the angle of rotation), which becomes non-zero at $T < T_c$, grows as $|Q| \propto |t|^\beta$, with the critical exponent β. Fifty years ago, Wilson showed that at T very close to $T_c$ the short-length details can be eliminated on scales below $1/e^\ell$ ($\ell$ counts the number of iterations in the elimination process), and that rescaling the unit length by the factor $e^\ell$ yields a renormalized effective (dimensionless) Hamiltonian $\hat{H}(\ell)$, which ‘flows’ in the space spanned by all such Hamiltonians. Often this renormalization group (RG) ‘flow’ reaches the vicinity of an $\ell$-independent fixed point (FP), $\hat{H}^*$. Adding small deviations from $\hat{H}^*$, $\hat{H} = \hat{H}^* + \sum \mu_i O_i$, with ‘operators’ $O_i$ and ‘coupling constants’ (or ‘scaling fields’) $\mu_i$, these scaling fields renormalize (to linear order) as $\mu_i(\ell) \approx e^{\lambda_i \ell} \mu_i(0)$. A scaling field $\mu_i$ is ‘relevant’ or ‘irrelevant’ when $\lambda_i > 0$ or $\lambda_i < 0$. The first two fields, which are always relevant, relate to $\mu_1(0) = t$ and to the ‘ordering field’ $\mu_2(0) = h$. $\mu_2(0) = h$.

Scaling.– After $\ell = \ell_f$ iterations, the whole correlation volume reduces to one unit cell, i.e., $\xi(\ell_f) = \xi_0 |t(\ell_f)|^{-\nu} = 1$, all fluctuations have been eliminated and simple techniques can be used to find the solution. Using $t(\ell_f) = e^{h\xi(\ell_f)} t$, this implies that $|t(\ell_f)|$ is also of order 1, and that $\lambda_1 = 1/\nu$. The singular part of the corresponding free-energy density then obeys the homogeneous scaling form

$$\mathcal{F}(\mu_i) = e^{-d\ell} \mathcal{F}(\mu_i(0) e^{\lambda_i \ell_f}) \equiv |t|^d W[h|t|^{-\nu\lambda_2}, \mu_3(0)|t|^{-\nu\lambda_3}, ...],$$

where $d$ is the dimensionality. Derivatives of $\mathcal{F}$ w.r.t. $t$ and $h$ yield the critical exponents for the measurable quantities, e.g. $\beta = \nu(d - \lambda_2)$. The exponents and the scaling function $W$ for systems in the corresponding universality class are fully determined by the FP itself, and not by the ‘initial’ effective Hamiltonian, which encompasses the short-scales behavior. Therefore they are ‘universal’.

For a ‘simple’ critical point, the other fields ($\mu_3, \mu_4, ...$) are irrelevant, $\lambda_3, \lambda_4, ... < 0$, decaying to zero, and the FP is ‘fully stable’. At finite $|t|$ the irrelevant fields only generate ‘corrections’ to the leading critical behavior, coming from the dependence of $W$ on the small $\mu_3(0)|t|^{-\nu\lambda_3}, \mu_4(0)|t|^{-\nu\lambda_4}, ...$. In some cases, a (third) field, $\mu_3$, is also relevant ($\lambda_3 > 0$), yielding an RG ‘flow’ away from the FP, which is then ‘unstable’. Such a FP represents a ‘multicritical point’; it can be reached only if the relevant field, $\mu_3(0)$, is tuned to zero. The outcome depends on ‘initial’ parameters: the flow can go to another, ‘stable’ FP, characterizing a distinct universality class (resulting with a ‘crossover’ between these two critical behaviors), or the flow does not approach any alternative FP, and $\hat{H}(\ell_f)$ may yield a ‘fluctuation-driven’ first-order transition. So far, there was only a limited discussion on the universality classes of such fluctuation-driven first-order transitions. Below we associate such transitions with new universality classes, and discuss the critical behavior as they are approached.

Universality.– Wilson’s theory derived the FP’s from cal-
calculations at $T > T_c$, and found a limited number of different FP’s, identified by the guiding paradigm: given the interaction range (in the microscopic Hamiltonian which describes the system), the dimensionality $d$ and the number of components of the order-parameter, $n$, the universality class of a FP is determined only by the symmetry above the transition, $T > T_c$. In the case of cubic crystals, there exist only two FP’s which can be ‘fully stable’, the ‘isotropic’ and the ‘cubic’ FP’s (see below). Theoretically, it was found that the isotropic (cubic) FP is stable for $n < n_c$, the value of the number of components of the order-parameter, $n$, the universality class of a FP is determined only by the symmetry above the transition, $T > T_c$. In contrast, the cubic to tetragonal transition first (at intermediate $|t|$’s) exhibits critical exponents from the vicinity of the unstable ‘isotropic’ FP, but eventually (close to $T_c$) undergoes a ‘fluctuation-driven’ first-order transition. The latter behavior applies to all the cubic to tetragonal transitions, and can thus be associated with a separate universality class. We suggest that other fluctuation-driven first-order transitions can also be identified by universality classes, associated with their symmetry below $T_c$. This suggestion, and the splitting of universal critical phenomena into separate low-$T$ symmetry determined universality classes, is our main result.

Numerically, the two competing FP’s at $n = d = 3$ are close, and it would seem difficult to differentiate between them. This may explain the false apparent universality between the two kinds of perovskites, seen in some experiments. Below we show that the RG flows in the vicinity of both FP’s are slow, and therefore they exhibit ‘effective’ exponents (which depend on the range of $t$, see below). We propose dedicated measurements of these exponents to test the different critical behaviors predicted here, and list examples of other phase transitions which may require similar renewed analyses.

The Wilson-Fisher Renormalization group. Ignoring fluctuations, phase transitions are described by a Landau expansion of the free-energy density in powers of the (small) order parameter components $Q_i$. The terms in this expansion are based on the symmetries of the system above the transition. For the isotropic $n$-component order parameter vector $Q$, this free-energy is $U_0(Q) = r|Q|^2/2 + u|Q|^4 + O(|Q|^6)$, where $r > 0$ and $u$ is a system-dependent parameter. In the cubic case this should be supplemented by $U_v(Q) = v \sum_{i=1}^{n} Q_i^4$, with $v$...
a system-dependent coefficient $^{10,27}$. Long wave-length fluctuations in $Q(r)$ are introduced via a gradient term, $|\nabla Q(r)|^2$, whose coefficient is normalized to 1. The effective Hamiltonian is then written as $\int d^d\tau \mathcal{H}(r)$, where

$$\mathcal{H}(r) \equiv |\nabla Q(r)|^2/2 + U_0|Q(r)| + U_v|Q(r)|, \quad (2)$$

with $Q(r)$ and $\nabla Q(r)$ generating the ‘operators’ discussed above. For $v = 0$, the Wilson-Fisher RG at $d = 4 - \epsilon$ gives $^{3,28}$ two FP’s, one at $U_0^c = 0$, termed ‘Gaussian’, and the ‘isotropic’ FP, with $U_0^f(n) = O(\epsilon) > 0$. The Gaussian FP is unstable, so that $u > 0$ flows to the isotropic FP and $u < 0$ flows to a region with a first-order transition (stabilized by the sixth order terms). The Gaussian FP is thus identified as a ‘tricritical point’.

**Cubic systems.** The RG analysis of the cubic model, Eq. (2), for general $n$ and in dimension $d = 4 - \epsilon$ $^{29,30}$ yielded four FP’s of order $\epsilon$: the Gaussian (G), $u_0^c = v_0^c = 0$), isotropic (I, $v_0^c = 0$, $u_0^c > 0$), decoupled Ising (D, $u_0^c = 0$, $v_0^c > 0$, for which the different $Q_i$‘s decouple from each other), and ‘cubic’ FP’s. The location of the cubic FP, $(u_0^c, v_0^c)$, depends on $n$: for small (large) $n$, it is in the lower (upper) half plane, as shown on the left (right) panel of Fig. 2. This figure $^{9}$ has been reproduced by other authors, e.g., Refs. 22,23,25, using various methods, and and included in textbooks. It shows the ‘critical surface’ of the effective Hamiltonian in the $u-v$ plane, on which $|t| = 0$ and $\xi = \infty$. At a finite (but very small) $|t|$ the RG flow starts very close to this surface, and as the RG is iterated the flow stays close to the arrows in the figure. When the flow reaches the vicinity of a FP the system exhibits the critical exponents of that FP.

As seen in Fig. 2, the Gaussian FP is doubly unstable: both $u$ and $v$ are relevant in its vicinity. The decoupled FP is singly unstable, with $u$ relevant $^{9,33}$. As mentioned, the stability of the isotropic and cubic FP’s depends on the borderline value $n_c(3)$. It is clear that $2 < n_c(3) < 4$, but different approximations yielded conflicting answers to the question whether $n = d = 3$ is above or below $n_c(3)$ (for the history, see Refs. 17,21–25 and references therein). For instance, a third-order $\epsilon$–expansion gave $^{9} n_c(3) \approx 3.128$ at $\epsilon = 1$. The result $3 < n_c(d = 3)$ was also obtained by the scaling-field method $^{34}$. Accepting this result, the RG flows are those shown on the left panel of Fig. 2.

We now show that when $3 < n_c(d = 3)$, both families of perovskites belong the the same universality class. At $T < T_c$, $Q$ becomes non-zero. For the isotropic case, with $v = 0$, $Q$ can point in arbitrary directions. However, $U_v$ breaks this symmetry. It is minimized (at $\ell = \ell_f$) for $Q$ along a cubic axis (tetragonal) when $v < 0$ and for $Q$ along a cubic diagonal (trigonal) when $v > 0$. As can be seen from the trajectories in Fig. 2, the RG iterations preserve these symmetries and the sign of $v$ does not change under them. Therefore, we conclude that for the cubic to tetragonal (trigonal) transition, the ‘initial’ effective Hamiltonian must be in the lower (upper) half plane. For $n < n_c(3)$, the region of attraction of the isotropic FP (shaded area on the left panel in Fig. 2) contains flows with both signs of $v$, and therefore one would conclude that the two types of transition belong to the same universality class, of the ‘isotropic’ FP.

![Figure 2](image-url)

**FIG. 2:** (color online) Schematic flow diagram and FP’s for the cubic model, Eq. (2), adapted from Ref. 9. G=Gaussian, I=isotropic, D=Decoupled (Ising) and C=Cubic FP’s. S=initial point for SrTiO$_3$. L=initial point for LaAlO$_3$. The dashed lines represent the stability edges, $u+v=0$ (for $v<0$) and $u+v/n=0$ (for $v>0$), below which the free energy in Eq. (2) is stabilized by the terms of order $|Q|^4$, and the transitions are first-order. The shaded areas are the regions of attraction of the stable FP’s (I on left and C on right).

As opposed, four very accurate methods (Monte Carlo simulations of lattice $O(n)$ models, $^{21}$ six-loop recursion relations $^{22}$ at $d = 3$, the good old $\epsilon$-expansion $^{23}$ to order $\epsilon^6$ and the bootstrap method, which calculates exponents at any dimension $^{24}$) find $2.85 < n_c(3) < 3$. Therefore, the RG flows are as in the right panel in Fig. 2: the isotropic FP is unstable [with a small but positive exponent for the ‘flow’ of $v$, $0 < \lambda^i \leq 0.02$, see Eq. (1)], while the cubic FP has a small but positive FP value $v_0^c > 0$, and is fully stable (deviations of both $u$ and $v$ from it decay under iterations).

Without even looking at the specific numerical values of the locations of the FP’s, the right panel of Fig. 2 yields qualitative crucial consequences: since the cubic to trigonal and cubic to tetragonal transitions correspond to opposite signs of $v$, they have different flow trajectories. For the former, $v < 0$. If the initial $|v|$ is small, and if the initial $u$ is positive, so that $u+v > 0$, then the respective effective Hamiltonian (shown by the blue trajectory leaving S in Fig. 2) first ‘flows’ closer to the isotropic FP, and may then exhibit $t$–dependent effective exponents associated with that FP, but eventually it must turn downwards, and cross the stability line $u+v = 0$ – turning the transition fluctuation-driven first-order. As $\lambda^i$ is small at $n = d = 3$, $^{21–24}$ this flow is slow, so that the first-order transition will occur only close to $T_c$, with a small discontinuity. For larger initial $|v|$’s the transition becomes first-order at larger $|t|$. In contrast, the cubic to trigonal transition has $v > 0$, and therefore its Hamiltonian must flow to the stable cubic FP, resulting in a second-order transition with cubic exponents.

As mentioned above, the cubic and the isotropic FP’s...
are close to each other at $n = d = 3$. Indeed, the calculated ‘asymptotic’ critical exponents (expected only at very small $|t|$) are $\nu_I \approx 0.706$, $\nu_C \approx 0.700$, $\beta_I \approx 0.366$, $\beta_C \approx 0.368$. This closeness also implies that the stability exponent of the stable cubic FP, $\lambda_0^C \approx 0$ and that of the unstable isotropic FP, $\lambda_0^I \approx 0$, are small, indicating slow flows towards and away from these FPs. The experimental exponent should therefore be compared with ‘effective exponents’, e.g., $\beta_{\text{eff}} \equiv \partial \log |Q|/\partial \log |t|$, which depend on $|t|$. For $v > 0$, the cubic FP is stable, with two negative stability exponents, $\lambda_0^I < \lambda_0^C < 0$. Therefore $\beta_{\text{eff}}$ approaches the asymptotic $\beta_C$ slowly, with corrections of order $|t||\phi_0^C|$, where $\phi_0^C = \nu_C \lambda_0^C$ is small. For $v < 0$, $\beta_{\text{eff}}$ first approaches the isotropic FP value $\beta_I$ (with the relatively large exponent $|\phi_0^I|$), but then (for smaller $|t|$, i.e. larger $\ell$), it moves away from that value, and $|Q|$ has a discontinuity. We expect that dedicated experiments, deducing the effective exponents from various ranges of $|t|$, will corroborate these predictions.

**Experiments on the perovskites.**—Based on the mean-field region of the experiments, Müller et al. estimated the ‘initial’ Landau parameters to be $\{u, v, \lambda\} \approx \{1.91, -0.068\}$ for SrTiO$_3$ and $\{u, v\}_0 \approx 0.06 \pm 0.06, 0.68 \pm 0.06\}$ for LaAlO$_3$, all in cgs units divided by $10^4$. These rough values (which should be improved!) fit beautifully with our expectations: SrTiO$_3$ (S in Fig. 2) has a small negative $v$, and we predict that it should flow near the isotropic FP before turning slowly towards first-order, while LaAlO$_3$ (L in Fig. 2) should flow slowly towards the cubic FP. Indeed, we found no experiments showing first-order transitions into the trigonal phase, but several experiments found such transitions into the tetragonal phase. First, some experiments hint that SrTiO$_3$ may be close to a tricritical point. Also, both RbCaF$_3$ and KMnF$_3$ have first-order transitions, confirming our theoretical predictions. All these systems exhibit intermediate regions with effective critical exponents $\beta \approx 0.40 \pm 0.03, 1/3, 0.27, 0.17 \pm 0.02$ for SrTiO$_3$, KMnF$_3$, RbCaF$_3$, and NaNbO$_3$, respectively. The latter smaller values have also been attributed to the inter-plane weak correlations along the rotation axis.

**Uniaxial stress.**—Frequently experimenters apply a uniaxial stress, $g$, which ‘prefers’ ordering along a single direction (or perpendicular to it). Such a perturbation is relevant near both the isotropic and cubic FP’s, scaling as $g(\ell) = g(0)e^{\lambda_\ell}$ with $\lambda_\ell > 0$ and yielding a crossover to a uniaxial or a transverse ordering. The scaling function $W$ in Eq. (1) also depends on $g/|t|^\phi_0$, with the ‘uniaxial anisotropy exponent’ $\phi_g = \nu \lambda_\ell$. As $|t|$ decreases, $W(g/|t|^\phi_0)$ becomes singular at some constant parameter $g/|t|^\phi_0 = y_0$, indicating the lower symmetry transition when $T_c(g) - T_c(0) \sim g^{1/\phi_0}$. Experiments with stress along the ‘easy axes’ on SrTiO$_3$ and [111] on LaAlO$_3$ were fitted to this form with $\phi_g = 1.27 \pm 0.06$ and $\phi_g = 1.31 \pm 0.07$, in apparent agreement with the ‘joint’ asymptotic isotropic-cubic value 1.27. Similar values of $\phi_g$ also came from elastic measurements: since $F = |t|^d W(g/|t|^\phi_0)$, the strains scale as $e_{ij} \sim |t|^{d - \phi_0}$, and the elastic compliance diverges as $|t|^{d - 2\phi_0}$.

**Mixed crystals.**—One way to vary $v$ experimentally is to use mixed crystals, e.g., Sr$_{1-x}$Ca$_x$TiO$_3$ or a mixture of SrTiO$_3$ with LaAlO$_3$, which should be easy to grow due to their matching lattice constants. Since both the isotropic and cubic FPs have $dv > 2$, randomness is irrelevant and one should see the same competition predicted above.

**Other $n = 3$ systems.**—Three-component order parameter vectors $Q$ with cubic symmetry are abundant. Examples include ferroelectric transitions from cubic to tetragonal or trigonal, ferromagnets and antiferromagnets with the magnetization ordering along an axis or a diagonal. In particular, our results should apply to the uniaxial antiferromagnets in a parallel magnetic field which ‘cancels’ the anisotropy to yield a (cubic or isotropic) point with $n = 3$. Close to their transitions, these cubic crystals may be described by Eq. (2), and therefore their critical behavior should split into the two universality classes described above. In particular, ordering of the $n = 3$ cubic system along a cube axis must be fluctuation-driven first-order, with intermediate effective isotropic exponents! This important prediction should be tested experimentally.

**Competing order parameters.**—Systems having two competing order parameters, $Q_1$ and $Q_2$, with $n_1$ and $n_2$ components (e.g., an Ising ferromagnet ($n_1 = 1$) and an incommensurate spin density wave with a complex order parameter which can be described by an $n_2 = 2$ model, or an isotropic antiferromagnet ($n_1 = 3$) and a superconductor ($n_2 = 2$), with $n = n_1 + n_2$, are often described by replacing the quartic terms in Eq. (2) by $U_3 = u_1|Q_1|^4 + u_2|Q_2|^4 + 2w|Q_1||Q_2|^2$. As before, the isotropic FP $u_1 = u_2 = w$ is stable for $n < n_c$. New accurate calculations show that for $n = 3$ this FP is unstable, and another FP (called ‘biconical’, with all three coefficients being non-zero) becomes stable. The flow diagram is similar to the right panel in Fig. 2: Hamiltonians with $w^2 \geq u_1 u_2$, which have separate ordered phases of $Q_1$ or of $Q_2$, flow to the vicinity of the isotropic FP, and then yield first-order transitions. On the other hand, Hamiltonians with $w^2 < u_1 u_2$ flow to the biconical FP (which replaces the cubic FP in our discussion). As in our cubic example, we predict that initial Hamiltonians on the two sides of the isotropic FP line belong to different universality classes. In contrast, for $n \geq 4$ there exists an exact scaling argument which shows that the stable fixed point is always decoupled, i.e., $w_{\text{eff}} = 0$. For modern examples, see Ref. 59. We shall elaborate on all these examples in a separate paper.

**Conclusions.**—In conclusion, we have shown the breakdown of the assumption that universality is determined.
by the symmetry above the transition. We also identified new universality classes, which depend on the symmetry below the transition, but then have a fluctuation-driven first-order transition. This resolves a long standing confusion about the displacive phase transitions in the perovskites, and leaves its complete confirmation to future dedicated experiments, concentrating on the $t$–dependence of the effective exponents. We expect similar phenomena in other cases with competing fixed points. As we showed, symmetry below the transition (e.g., tetragonal) is sufficient to predict a fluctuation-driven first-order transition.

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At $t = 0$, $ξ = ∞$, and the flow stays on the $u − v$ plane. For a finite sample, the cutoff $ξ$ in Eq. (1) is replaced by the system’s linear size, $L$. Macroscopic quantities, like the average order-parameter, are then powers of $L$, e.g. $⟨Q⟩ ∝ L^{-3/6}$. This is the basis for finite-size scaling.

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