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Random-Field-like Criticality in Glass-Forming Liquids

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We introduce an approach to derive an effective scalar field theory for the glass transition; the fluctuating field is the overlap between equilibrium configurations. We apply it to the case of constrained liquids for which the introduction of a conjugate source to the overlap field was predicted to lead to an equilibrium critical point. We show that the long-distance physics in the vicinity of this critical point is in the same universality class as that of a paradigmatic disordered model: the random-field Ising model. The quenched disorder is provided here by a reference equilibrium liquid configuration. We discuss to what extent this field-theoretical description and the mapping to the random field Ising model hold in the whole supercooled liquid regime, in particular, near the glass transition.

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Introduction.—One of the first steps in the analysis of a standard phase transition consists of identifying the correct order parameter. Once this crucial step is done, one can first construct a Landau functional and analyze it to obtain a mean-field description; then, eventually, one can promote the order parameter to a truly fluctuating field and study the associated field theory in order to get a full-fledged theoretical description. In the case of the glass transition, establishing what is the correct order parameter is by no means an easy task. The random first-order transition (RFOT) theory [1,2] identifies as order parameter the similarity, also called overlap, between equilibrium configurations. More specifically, one should take an equilibrium (reference) configuration and then restrict the thermodynamic sampling only to equilibrium configurations constrained to have a high overlap with the reference one on the boundary of the sample. By definition, a RFOT takes place when this boundary condition forces the entire system to have a large value of the overlap instead of the zero (or very low) one characteristic of the liquid phase. This is analogous to forcing a positive boundary magnetization in the case of a ferromagnetic phase transition. The identification of this order parameter was first made in the context of disordered mean-field systems [3], but it was soon realized that the overlap and its fluctuations provide an interesting tool in the study of glassy systems, in particular, supercooled liquids, irrespective of the presence or not of quenched disorder [4]. Studying the response to perturbations directly acting on the order parameter, as done in usual phase transitions, has recently allowed access to the growing static length accompanying the slowing down of the dynamics. In the RFOT context, the perturbation is a pinning field that forces configurations to have a high overlap locally; the corresponding correlation length is called "point to set" [5]. As discussed before, promoting the order parameter to a fully fluctuating field is a way to study fluctuations and correlations beyond mean-field theory: this was recently done to analyze the slow but intermediate β relaxation in the vicinity of the "ideal" dynamical transition that is found both in the mode-coupling approach and in the RFOT theory of liquids [6,7]. In particular, it has been shown that the dynamical transition is in the universality class of the spinodal of the random-field Ising model (RFIM). Both singularities, of course, can only be present when activated events such as nucleation are not taken into account. Finally, the overlap has also been the focus of an intense numerical research in model supercooled liquids: the distribution of the fluctuations of the uniform overlap between equilibrium configurations has been computed and found to develop a nontrivial, non-Gaussian shape as one cools the liquid [8–10].

The aim of our work is to develop an effective field theory of glass-forming systems directly formulated in terms of an overlap field. This is highly desirable for several reasons: first, it allows one to focus directly on what is thought to be the physically relevant field; second, it leads to a scalar field theory in the presence of quenched disorder and should, therefore, settle the recurrent debate about whether the glass transition is related to random-field, random-bond, or spin-glass physics [2,11,12]. Our approach is able to capture nonperturbative effects conjectured to be crucial to describe the glass transition: by

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perturbatively integrating out irrelevant degrees of freedom only, we derive an effective theory for the relevant field, identified as the overlap with a reference equilibrium configuration. The model (or field theory) obtained by this procedure can then be nonperturbatively studied either by renormalization-group analysis or by computer simulations.

In the following, we first introduce our method in a general setting for glass-forming liquids. Then, we apply it to the *critical point* that terminates the transition line in an extended phase diagram where one introduces a coupling between liquid configurations, and we show that the critical behavior is the same as in the equilibrium RFIM. The motivation for studying this specific region of parameters, besides providing a first progress towards a comprehensive field theory of the glass transition, stems from recent numerical works [8–10] that have directly focused on the behavior of supercooled liquids in the presence of such an attractive coupling and have provided evidence for a first-order transition line and a terminal critical point. In consequence, our predictions are prone to direct tests in the future. (Note that the analysis of Ref. [13] does not apply here since, in the presence of a nonzero coupling, the first-order transition line is *not* a RFOT and the spinodal is *not* equivalent to a dynamical mode-coupling-like transition [14].)

Consider a glass-forming liquid formed by N particles and described by a Hamiltonian $H[\mathbf{r^N}]$ where $\mathbf{r^N}$ denotes a configuration of the N particles. We consider a reference equilibrium configuration $\mathbf{r_0^N}$ and define the overlap at point x between the latter and another configuration $\mathbf{r^N}$ as $\hat{q}_x[\mathbf{r^N},\mathbf{r_0^N}] := \int_{\mathbf{y}} \mathbf{f}(\mathbf{y})[\hat{\rho}(\mathbf{x}+(\mathbf{y}/\mathbf{2})|\mathbf{r^N})\hat{\rho}(\mathbf{x}-(\mathbf{y}/\mathbf{2})|\mathbf{r_0^N})-\rho^{\mathbf{2}}]$, where $\int_y \equiv \int d^D y$ and $f(y)$ is a smoothing function of short range (typically the cage size); $\rho(x|\mathbf{r^N}) = \sum_{\mathbf{i=1}}^{\mathbf{N}} \delta^{(D)}(\mathbf{x}-\mathbf{r_i})$ is the microscopic density at point x, and ρ is the mean liquid density. We can now define an overlap field $p(x)$ and introduce an effective Hamiltonian or action for this field,

$$S[p|\mathbf{r_0^N}] = -\log \int \frac{d\mathbf{r^N}}{N!} \delta[p - \hat{q}[\mathbf{r^N},\mathbf{r_0^N}]] e^{-\beta H[\mathbf{r^N}]}, \qquad (1)$$

where $\delta[]$ is a functional that enforces a delta function at each point x and $\beta = 1/(k_B T)$. The probability to observe a certain profile of the overlap field is given by $\exp(-S[p|\mathbf{r_0^N}])$. Thermodynamic quantities and correlation lengths, e.g., point-to-set ones, are obtained as usual from a "partition function" and the associated functional W,

$$e^{W[\epsilon|\mathbf{r_0^N}]} = \int \mathcal{D}p e^{-S[p|\mathbf{r_0^N}] + \int d^D x \epsilon(x) p(x)}, \qquad (2)$$

where we have introduced an auxiliary coupling $\epsilon(x)$ that, in field-theoretical language, plays the role of a "source" for generating the connected correlation functions of the overlap field. A RFOT corresponds to the appearance for $\epsilon = 0$ of long-range order in the overlap field, which acquires a large value in the entire sample. Because of the reference configuration $\mathbf{r_0^N}$, the action S describes a scalar field theory in the presence of quenched disorder. In order to analyze it and understand in more detail what kind of disorder is generated by $\mathbf{r_0^N}$, one can study the cumulants of S by considering replicas of the original system. As known in the context of the critical behavior of the RFIM [15,16] (see also Refs. [6,17]), $\exp(-S_{\rm rep}[\{p_a\}]) = \overline{\exp(-\sum_{a=1}^n S[p_a|\mathbf{r_0^N}])}$ generates the cumulants of the action $S[p|\mathbf{r_0^N}]$ through an expansion in increasing number of free replica sums [15,16]

$$S_{\rm rep}[\{p_a\}] = \sum_{a=1}^n S_1[p_a] - \frac{1}{2}\sum_{a,b=1}^n S_2[p_a,p_b] + \frac{1}{3!}\sum_{abc=1}^n S_3[p_a,p_b,p_c] + \cdots, \qquad (3)$$

where $S_l[p_1,\ldots,p_l]$ is the lth cumulant: e.g., $\overline{S[p|\mathbf{r_0^N}]} = S_1[p]$ and $\overline{S[p_1|\mathbf{r_0^N}]S[p_2|\mathbf{r_0^N}]} - \overline{S[p_1|\mathbf{r_0^N}]}\,\overline{S[p_2|\mathbf{r_0^N}]} = S_2[p_1,p_2]$. The Franz-Parisi potential [3], which is the average free-energy cost to keep two configurations at a fixed global overlap, is the Legendre transform of the first cumulant of $W[\epsilon|\mathbf{r_0^N}]$ (and is not equal to S_1 except in the mean-field limit).

Our goal is to derive the action for the overlap field and its cumulants in glass-forming systems. To proceed, one can formally rewrite

$$e^{-S_{\rm rep}[\{p_a\}]} \propto \int d\mathbf{r_0^N} \prod_{a=1}^n d\mathbf{r_a^N} \delta[p_a - \hat{q}[\mathbf{r_a^N},\mathbf{r_0^N}]] \times e^{-\beta(H[\mathbf{r_0^N}] + \sum_{a=1}^n H[\mathbf{r_a^N}])}. \qquad (4)$$

The n replicas plus the reference configuration can now be described by greek letters $\alpha = 0,1,\ldots,n$ whereas roman letters are still used for replicas from 1 to n only. We also introduce additional collective fields $q_{\alpha\beta}(x)$ that describe the overlap between two different replicas α and β. Equation (4) then becomes

$$e^{-S_{\rm rep}[\{p_a\}]} \propto \int \prod_{ab\neq} Dq_{ab} \left[\int \prod_{\alpha} d\mathbf{r_\alpha^N} \prod_{\alpha\beta\neq} \delta[q_{\alpha\beta} - \hat{q}[\mathbf{r_\alpha^N},\mathbf{r_\beta^N}]] \times e^{-\beta\sum_\alpha H[\mathbf{r_\alpha^N}]}\right] \propto \int \prod_{ab\neq} Dq_{ab} e^{-S[\{p_a,q_{ab}\}]}, \qquad (5)$$

where we have used the notation $q_{0a} = q_{a0} \equiv p_a$ and $S[\{p_a,q_{ab}\}]$ is defined as minus the logarithm of the expression between square brackets. Our approach differs from the usual replica one in that it treats the fields $p_a = q_{0a}$ and q_{ab} differently. The underlying working hypothesis, which can at least be checked in the vicinity of the terminal critical point in the presence of a nonzero coupling

ϵ, is that the p_a's may develop long-range fluctuations while, for fixed p_a's, the q_{ab}'s are harmless for the long-distance physics and can be approximately integrated out. In the following we show how this procedure can be carried out near the terminal critical point.

One first needs a tractable expression for the action $S[\{p_a, q_{ab}\}]$ in terms of the overlap fields [defined via Eq. (5)]. This action of course depends on the microscopic details of the glass-forming system under study, and deriving its expression can be a rather formidable task. One can formally derive $S[\{q_{\alpha\beta}\}]$ from the Morita-Hiroike functional of the 1- and 2-particle densities [18] of the replicated $(n+1)$-component liquid mixture along lines similar to those followed by Ref. [7]. A short cut is, however, provided by the coarse-grained effective Landau-like functional considered in Ref. [17],

$$S[\{q_{\alpha\beta}\}] = \frac{E_0}{k_B T}\int_x \left\{\frac{c}{2}\sum_{\alpha\beta\neq}(\partial q_{\alpha\beta}(x))^2 + \sum_{\alpha\beta\neq} V(q_{\alpha\beta}(x)) - \frac{u}{3}\sum_{\alpha\beta\gamma\neq} q_{\alpha\beta}(x) q_{\beta\gamma}(x) q_{\gamma\alpha}(x)\right\}, \tag{6}$$

where $V(q) = (t/2)q^2 - [(u+w)/3]q^3 + (y/4)q^4$ and the primary dependence on temperature is given by $t \approx k_B(T - T_0)/E_0$ with E_0 the typical energy scale of the liquid and T_0 a constant with dimension of temperature. In numerical applications, we focus on the parameter values found to roughly reproduce properties of glass-forming liquid ortho-terphenyl: $u = 0.385$, $w = 2.73$, $y = 1.82$, and $c = 1$ (in appropriate length units) [17].

To derive an effective field theory for the overlaps $p_a(x)$ with a reference configuration, one needs to perform the functional integration over the $q_{ab}(x)$'s while keeping the fields $p_a(x)$ fixed [see Eq. (5)]. It is clear from Eq. (6) that, as discussed above, nonzero p_a's exert an external source or field on the q_{ab}'s. In Eq. (6) the cubic term generates a contribution $-u\sum_{ab\neq} p_a(x) p_b(x) q_{ab}(x)$. In consequence, the q_{ab}'s do not develop fluctuations on all scales and stay "massive" near the terminal critical point [19]. It is then sufficient to perform the functional integration on these fields through a perturbative treatment. Perturbation is carried out with a saddle-point approximation as zeroth order. The saddle-point equations for the q_{ab}'s with $a \neq b$ read

$$c\partial^2 q_{ab*}(x) + V'(q_{ab*}(x)) = u p_a(x) p_b(x) + u\sum_{c\neq a,b} q_{ac*}(x) q_{cb^*}(x). \tag{7}$$

The solution of this equation has to be inserted back into the action in order to obtain the final result. Since we are interested in the long wavelength fluctuations of the p_a's, it is sufficient to solve the saddle-point equations in an expansion in the gradient term, the zeroth order then corresponding to simply neglecting the gradient.

All quantities can be expanded in increasing number of free replica sums as in Eq. (3), e.g.,

$$q_{ab*}(x) = q_x^{[0]}[p_a, p_b] + \sum_c q_x^{[1]}[p_a, p_b | p_c] + O\left(\sum_{cd}\right). \tag{8}$$

Such expansions allow algebraic manipulations that lead to well defined and unique expressions of the various orders [15,16,20]. In the lowest order in the gradient amplitude parameter c, Eqs. (7) and (8) lead, for instance, to $q_x^{[0]}[p_a, p_b] \equiv q^{[0]}(p_a(x), p_b(x))$ and

$$V'(q^{[0]}(p_a, p_b)) = u p_a p_b - u q^{[0]}(p_a, p_b) \times [q^{[0]}(p_a, p_a) + q^{[0]}(p_b, p_b)]. \tag{9}$$

With the above results, one immediately derives from Eqs. (3) and (5) the expressions of the first two cumulants of the action for the p_a's at the level of the saddle-point approximation and including $O(\partial^2)$ terms only

$$S_1[p_a] = \frac{E_0}{k_B T}\int_x \left\{c(\partial p_a)^2 - \frac{1}{2}c(\partial q^{[0]}(p_a, p_a))^2 + 2V(p_a) - V(q^{[0]}(p_a, p_a)) - \frac{2u}{3} q^{[0]}(p_a, p_a)^3 + u p_a^2 q^{[0]}(p_a, p_a)\right\} \tag{10}$$

and

$$S_2[p_a, p_b] = -\frac{E_0}{k_B T}\int_x \left\{\frac{c}{2}(\partial q^{[0]}(p_a, p_b))^2 - 2u p_a p_b q^{[0]}(p_a, p_b) + u[q^{[0]}(p_a, p_a) + q^{[0]}(p_b, p_b)] q^{[0]}(p_a, p_b)^2 + 2V(q^{[0]}(p_a, p_b))\right\}, \tag{11}$$

where the explicit x dependence has been omitted. This derivation is easily extended to the higher orders, but the algebra rapidly becomes tedious, and the results are given in the Supplemental Material [21].

The above cumulants describe a scalar field theory for a disordered system [6,15,16,20]. We now more specifically consider the vicinity of the terminal critical point in the (T, ϵ) plane. At the saddle-point level, the critical point is defined by the following conditions:

$$\left.\frac{\partial S_1(p_1)}{\partial p_1}\right|_c = \epsilon_c, \quad \left.\frac{\partial^2 S_1(p_1)}{\partial p_1^2}\right|_c = 0, \quad \left.\frac{\partial^3 S_1(p_1)}{\partial p_1^3}\right|_c = 0. \tag{12}$$

When inserted in Eq. (10), this gives $\epsilon_c = 0.602E_0/(k_B T_c)$, $t_c = 1.680$, $p_c = 0.534$, and $q_c^{[0]} = 0.072$ (using the relation [17] between t and T one finds $E_0/(k_B T_c) = 0.952$). One can then expand the cumulants $S_l[p_1, \ldots, p_l]$ around the critical value after defining $\phi_a(x) = p_a(x) - p_c$. Keeping only terms to order ϕ^4 (higher-order terms are expected to be irrelevant at criticality) leads to a Wilson-Ginzburg-Landau action for the replica scalar fields $\phi_a(x)$

$$\begin{aligned} S_{\rm rep}[\{\phi_a\}] - \epsilon_c \int_x \phi_a(x) = \int_x \Bigg\{ & \sum_a \left[c'(\partial\phi_a)^2 + \frac{r_2}{2}\phi_a^2 + \frac{r_3}{3!}\phi_a^3 + \frac{r_4}{4!}\phi_a^4\right] \\ & - \frac{1}{2}\sum_{ab}\phi_a\phi_b\left[\Delta_{20} + \frac{\Delta_{21}}{2}(\phi_a + \phi_b) + \frac{\Delta_{22}}{4}\phi_a\phi_b + \frac{\Delta_{23}}{2}(\phi_a^2 + \phi_b^2)\right] \\ & + \frac{1}{3!}\sum_{abc}\phi_a\phi_b\phi_c\left[\Delta_{30} + \frac{\Delta_{31}}{3}(\phi_a + \phi_b + \phi_c)\right] - \frac{\Delta_{40}}{4!}\sum_{abcd}\phi_a\phi_b\phi_c\phi_d + O\left(\sum_{abcde}\right)\Bigg\}, \end{aligned} \tag{13}$$

where r_2 and r_3 are zero at the saddle-point (mean-field) critical point and all other coefficients are evaluated at the latter point: $c' = 0.91$, $r_4 = 23.33$, $\Delta_{20} = 0.22$, $\Delta_{21} = 0.47$, $\Delta_{22} = 1.23$, $\Delta_{23} = 0.04$, $\Delta_{30} = -0.12$, $\Delta_{31} = -0.51$, $\Delta_{40} = 0.33$. We have neglected some square gradient terms of higher orders in the fields because they are either less relevant in the renormalization group (RG) sense or are anyhow generated along the RG flow. Beyond the precise values, the important fact is that r_4, Δ_{20}, Δ_{22}, and Δ_{40} are greater than zero. It is then easily inferred that the above replicated action is obtained from a random one $S[\phi|h, \delta r_2, \delta r_3]$, which describes the large-scale fluctuations of the overlap field $p(x)$ with a reference equilibrium configuration

$$\begin{aligned} S[\phi|h, \delta r_2, \delta r_3] = \int_x \Bigg\{ & c'[\partial\phi(x)]^2 + \frac{r_2}{2}\phi(x)^2 + \frac{r_3}{3!}\phi(x)^3 \\ & + \frac{r_4}{4!}\phi(x)^4\Bigg\} + \int_x \Bigg\{\frac{\delta r_2(x)}{2}\phi(x)^2 \\ & + \frac{\delta r_3(x)}{3!}\phi(x)^3 - h(x)\phi(x)\Bigg\}, \end{aligned}$$

where the random field $h(x)$, the random mass $\delta r_2(x)$, and the random cubic coupling $\delta r_3(x)$ are delta-correlated processes with zero mean and higher cumulants given by

$$\begin{aligned} \overline{h(x)h(x')} &= \delta_{xx'}\Delta_{20}, \\ \overline{h(x)h(x')h(x'')} &= \delta_{xx'x''}\Delta_{30}, \\ \overline{h(x)h(x')h(x'')h(x''')}|_{\rm cum} &= \delta_{xx'x''x'''}\Delta_{40}, \\ \overline{\delta r_2(x)\delta r_2(x')} &= \delta_{xx'}\Delta_{22}, \\ \overline{h(x)\delta r_2(x')} &= -\delta_{xx'}\Delta_{21}, \\ \overline{h(x)h(x')\delta r_2(x'')} &= \delta_{xx'x''}\Delta_{31}, \\ \overline{h(x)\delta r_3(x')} &= -\delta_{xx'}\Delta_{23}, \end{aligned}$$

etc., with $\delta_{xx'}, \delta_{xx'x''}, \ldots$ short-hand notations for delta functions and products of delta functions. The resulting theory is, thus, a ϕ^4 one in the presence of quenched disorder without statistical inversion (Z_2) symmetry. As is known from the theory of disordered systems [24], the random field is the most relevant of the above terms at criticality and it leads to a universality class controlled by a nontrivial zero-temperature fixed point. This is so even in the absence of Z_2 symmetry, as can be found from a simple Harris-like criterion and has recently been confirmed by a full nonperturbative RG analysis [25].

The conclusion of our analysis is that the terminal critical point found in the presence of a conjugate source within the mean-field theory belongs in finite dimension to the universality class of the RFIM. Of course this is valid *if the transition is not destroyed by the disorder.* One, therefore, needs to compare the "bare" strength of the random field $\sqrt{\Delta}$ obtained from S_2 to that of the surface tension Y obtained from S_1. We have computed the latter as the free-energy cost per unit surface between two regions with high and low overlap [26,27] at coexistence far below the (mean-field) critical point and evaluated Δ at the same temperature (see the Supplemental Material [21]). The output is $\sqrt{\Delta}/Y \simeq 0.47$, which from known numerical results on the RFIM in $d = 3$ is compatible (Supplemental Material [21]) with the existence of a transition [24]. We have repeated the whole analysis for the finite dimensional 3-spin model with weak long-range interactions [21] and obtained that the mapping to the RFIM also holds. However, in this case, we have found $\sqrt{\Delta}/Y \simeq 2.24$, a value likely too large for a transition to survive in $d = 3$. As a result, no glass transition (RFOT) is expected in the model, in agreement with results and arguments presented in Refs. [28,29].

The Landau-like functional for glass-forming liquids [11,17] used as initial input in the present field-theoretical approach is just a crude approximation. Deriving from first principles a proper starting point with effective parameters that incorporate the microscopic information about glass formers is then a crucial task. As illustrated by recent numerical work [9,10], this is now within reach. Indeed, by constraining the overlap to a fixed value in a *small-size*

system (less than the point-to-set length), it should be possible to measure the local part of the first cumulants S_1 and S_2 (see above), thus allowing a direct evaluation of the *bare magnitude* of the different sources of quenched disorder in model supercooled liquids.

An important question for the theory of glass-forming systems is to what extent the mapping to the RFIM found near the terminal critical point in the presence of a nonzero coupling ϵ is general? As a first piece of an answer, we have verified that it also holds for the transition line in the temperature $\epsilon > 0$ plane, which is, therefore, a first-order transition in the presence of a random field [30]. When approaching the situation in the absence of coupling, $\epsilon = 0$, where an ideal glass transition of RFOT type is predicted at the mean-field level, establishing the mapping is not straightforward. It can be done for completely connected models (one can, e.g., show that the random energy model [31] maps exactly to a zero-dimensional RFIM). However, an extension to finite-dimensional systems is nontrivial as there may be long-wavelength fluctuations associated with diverging point-to-set spatial correlations. This will be the focus of a future publication.

After completion of our work we came to know that S. Franz and G. Parisi have also addressed the problem of the critical point of constrained glassy systems [33]. Their approach is different but leads to the same conclusion regarding the universality class of the terminal point in the T-ϵ plane. We acknowledge support from the ERC Grants No. NPRGGLASS and (C. C.) CRIPHERASY (No. 247328).

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