Stripe Glasses: Self-Generated Randomness in a Uniformly Frustrated System

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We show that a system with competing interactions on different length scales, relevant to the formation of stripes in doped Mott insulators, undergoes a self-generated glass transition which is caused by the frustrated nature of the interactions and not related to the presence of quenched disorder. An exponentially large number of metastable configurations is found, leading to a slow, landscape dominated long time relaxation and a breakup of the system into a disordered inhomogeneous state.

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Competing interactions on different length scales are able to stabilize mesoscale phase separations and the creation of spatial inhomogeneities in a wide variety of systems. Examples are stripe formation in doped Mott insulators, as found in transition metal oxides (TMO) [1,2], domains in magnetic multilayer compounds [3,4], or mesoscopic structures formed by assembling polymers in solution and amphiphiles in water-oil mixtures [5,6]. In many of these cases the tendency towards a perfectly ordered array of domains, stripes, etc. is undermined by frustrating long range interactions [7]. Very often, these assemblies exhibit a long time dynamics similar to the relaxation seen in glasses. In the context of stripes it has been argued that the presence of only very few quenched impurities might already cause a strictly disordered glassy state [8]. Furthermore, recent molecular dynamics calculations for charge ordering in TMO found an anomalous long time relaxation with a power spectrum similar to $1/f$ noise [9]. Indeed, there is experimental evidence for the formation of intrinsic inhomogeneities and even a stripe glass in high temperature superconductors and other transition metal oxides [10–17]. In particular slow, activated dynamics as observed in NMR experiments [13,15] exhibits a striking universality, rather independent of the details of added impurities, etc. It is therefore tempting to speculate that glassiness in these systems is *self-generated* and does not rely on the presence of quenched disorder, which may of course further stabilize a glassy state.

In this paper we show that the competition of interactions on different length scales in a uniformly frustrated system exhibits a self-generated glass transition due to the emergence of an exponentially large number of metastable states. This result is obtained by using the replica approach of Refs. [18,19] and by solving the corresponding many body problem using the self-consistent screening approximation [20,21]. Since only very few examples exist for models which exhibit self-generated glassiness [22,23], all these approaches are extremely important for a better understanding of glassiness in general. Even though our findings apply to a broader class of problems than stripes in TMO, we will adopt a language which is specific to that problem [24].

A model for a uniformly frustrated system with competition on different length scales is given by the Hamiltonian [7]

$$\mathcal{H} = \frac{1}{2}\int d^3x\left\{r_0\varphi(\mathbf{x})^2 + [\nabla\varphi(\mathbf{x})]^2 + \frac{u}{2}\varphi(\mathbf{x})^4\right\} + \frac{Q}{2}\int d^3x \int d^3x' \frac{\varphi(\mathbf{x})\varphi(\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|}. \quad (1)$$

Here, $\varphi(\mathbf{x})$ characterizes charge degrees of freedom, with $\varphi(\mathbf{x}) > 0$ in a hole-rich region, $\varphi(\mathbf{x}) < 0$ in a hole-poor region, and $\varphi(\mathbf{x}) = 0$ if the local density equals the averaged one. If $r_0 < 0$ the system tends to phase separate since we have to guarantee charge neutrality, $\langle\varphi\rangle = 0$. The coupling constant, Q, is a measure for the strength of the Coulomb interaction and characterizes the competition between short and long range interactions. In the case of strongly anisotropic, quasi–two-dimensional cuprate superconductors one expects an anisotropy of the gradient term in Eq. (1), which we neglect for simplicity. Despite the absence of a clean derivation of Eq. (1) from the many electron Schrödinger equation, we note that it describes, on a phenomenological level, many of the major competing effects which yield in microscopic theories a rich phase diagram of inhomogeneous spin and charge structures [25]. For $Q = 0$ and $r_0 < 0$ we expect at low temperatures long range ordered charge modulations. As shown in Ref. [26], the Coulomb interaction suppresses this ordered state for all $Q > 0$ and finite T. Instead, the system undergoes several crossovers. Most interestingly, at low temperatures, where $|r(T)| < 2\sqrt{Q}$, a mean field analysis of Eq. (1) shows that besides a correlation length, $\xi = 2(r + 2\sqrt{Q})^{-1/2}$, an additional length scale, $l_m = 4\pi(2\sqrt{Q} - r)^{-1/2}$, emerges, which characterizes the spatial modulation of the field correlations [26], where $r = r_0 + uT\langle\varphi^2\rangle$. These modulations are particularly relevant for low enough T where $r(T) \leq 0$, where mean field theory gives locally ordered regions with characteristic size $l_m \ll \xi$. We will show that a *stripe glass* emerges in this temperature regime.

An essential prerequisite for the anomalous dynamical features of glassiness, like aging, memory effects, and

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ergodicity breaking, is most certainly the occurrence of a large number of metastable states, $\mathcal{N}_{\rm ms}$, separated by energy barriers which are large compared to the temperature. In viscous liquids undergoing vitrification calorimetry suggests $\mathcal{N}_{\rm ms} \propto \exp(\text{const}V)$, where V is the system size. This observation is the heart of an *ideal glass transition* scenario based on random first order transitions [27], which was originally motivated by microscopic stability analyses of structural glasses and mean field theories for random Potts glasses. Below a crossover temperature, T_A, a "viscous," energy-landscape dominated long time relaxation sets in due to the occurrence of exponentially many metastable states, i.e., the configurational entropy, $S_c = k_B \log \mathcal{N}_{\rm ms}$, becomes extensive. Because of the large barriers between these states, the system will get stuck for extremely long times in one of the metastable states, i.e., it will freeze into a glass, at some temperature $T_G < T_A$ which depends, for example, on the cooling rate. Even though this laboratory glass transition is purely dynamical, a key ingredient of the ideal glass transition scenario is that the dynamical slowing arises from proximity to an underlying phase transition at $T_K < T_G$, where the configurational entropy would vanish like $S_c(T) \propto T - T_K$. If such an ideal transition exists, even for an infinitely slow cooling rate, freezing will occur at T_K since all the liquid degrees of freedom die out due to this "entropy crisis" [28].

Detailed theoretical investigation of this scenario has concentrated on systems with quenched randomness. A major step forward for studying nonrandom systems was made in Refs. [18,19], where a new replica approach was developed. Within this approach, the configurational entropy for a model of a structural glass without quenched disorder was calculated and found to be in good agreement with computer simulations [19].

We will use this approach to determine S_c for a system governed by Eq. (1). The key idea is to introduce, in analogy to the theory of conventional phase transitions, an appropriate symmetry breaking field $\psi(\mathbf{r})$, and to compute the partition sum

$$\tilde{Z}[\psi] = \int D\varphi\, e^{-\mathcal{H}[\varphi]/T - (g/2)\int d^3x[\psi(\mathbf{x})-\varphi(\mathbf{x})]^2}, \qquad (2)$$

where $g \to 0^+$. The energy $\tilde{f}[\psi] = -T \log \tilde{Z}[\psi]$ will be low if $\psi(\mathbf{r})$ equals to configurations which locally minimize $\mathcal{H}$. Sampling all configurations of the ψ field, weighted with $\exp(-\tilde{f}[\psi]/T)$, is therefore equivalent to scanning all metastable states such that

$$\tilde{F} = \lim_{g\to 0} \frac{1}{W} \int D\psi \tilde{f}[\psi] \exp(-\tilde{f}[\psi]/T) \qquad (3)$$

is a weighted average of the free energy in the various metastable configurations, where $W = \int D\psi \exp(-\tilde{f}[\psi]/T)$ is introduced for proper normalization. If there are only a few local minima, the limit $g \to 0^+$ behaves perturbatively and $\tilde{F}$ equals to the free energy, F, of the system. However, in case exponentially many local minima with large barriers between them exist, a nontrivial contribution arises from the ψ integral even for $g \to 0^+$, and the averaged free energy, $\tilde{F}$, differs from F. This enables us to identify the configurational entropy S_c via $F = \tilde{F} - TS_c$ [18,19]. For an illustration of the corresponding free energy landscape, see the inset of Fig. 1.

An explicit expression for S_c can be obtained within a replicated theory [18] with

$$F(m) = -\lim_{g\to 0} \frac{T}{m} \log \int D\psi \tilde{Z}^m[\psi]. \qquad (4)$$

It follows that $\tilde{F} = \frac{\partial m F(m)}{\partial m}|_{m=1}$, which gives

$$S_c = \frac{1}{T} \frac{\partial F(m)}{\partial m}\bigg|_{m=1}. \qquad (5)$$

Inserting $\tilde{Z}[\psi]$ of Eq. (2) into Eq. (4) finally leads to

$$Z(m) = \lim_{g\to 0} \int D^m\varphi \exp\left(-\sum_{a=1}^{m} \mathcal{H}[\varphi^a]/T - \frac{g}{2m} \sum_{a,b=1}^{m} \int d^3x \varphi^a(\mathbf{x})\varphi^b(\mathbf{x}) \right), \qquad (6)$$

with $F(m) = -\frac{T}{m}\log Z(m)$. Equation (6) has a formal similarity to the action of the random field Ising model, obtained within the conventional replica approach, which allows us to use techniques developed for that model [21]. In the following we use the self-consistent screening approximation (SCSA) of Eq. (6) [20,21,29] and determine the Green's function, $\mathcal{G}_{ab}(\mathbf{q}) = \langle \varphi^a(\mathbf{q})\varphi^b(-\mathbf{q})\rangle$, in replica space. $\mathcal{G}_{ab}(\mathbf{q})$ then determines the partition function $Z(m)$, and correspondingly S_c.

The interaction between different replicas is symmetric with respect to the replica index suggesting the mean field ansatz

$$\mathcal{G}_{ab}(\mathbf{q}) = [\mathcal{G}(\mathbf{q}) - \mathcal{F}(\mathbf{q})]\delta_{ab} + \mathcal{F}(\mathbf{q}), \qquad (7)$$

with equal diagonal elements $\mathcal{G}(\mathbf{q})$, and equal off-diagonal elements $\mathcal{F}(\mathbf{q})$ [30]. The physical interpretation of $\mathcal{G}(\mathbf{r} - \mathbf{r}') = \langle \varphi(\mathbf{r})\varphi(\mathbf{r}')\rangle$ as the thermodynamic (instantaneous) correlation function is straightforward. On the other hand, $\mathcal{F}(\mathbf{r} - \mathbf{r}') = \lim_{t\to\infty}\langle \varphi(\mathbf{r},t)\varphi(\mathbf{r}',0)\rangle$ can be interpreted as measuring long time correlations, arising from trapping in metastable minima which, in mean field theory, have infinite barriers between them. An analogous structure in replica space follows for the diagonal elements $\Sigma_{\mathcal{G}}(\mathbf{q})$, and off-diagonal elements $\Sigma_{\mathcal{F}}(\mathbf{q})$, of the self-energy, which are given in the SCSA as

$$\Sigma_{\mathcal{A}}(\mathbf{q}) = 2 \int \frac{d^3p}{(2\pi)^3} \mathcal{D}_{\mathcal{A}}(\mathbf{p})\mathcal{A}(\mathbf{p} + \mathbf{q}) \qquad (8)$$

with $\mathcal{A} \in \{\mathcal{G}, \mathcal{F}\}$. The screening of the interaction is characterized by the collective propagators $\mathcal{D}_{\mathcal{G}}^{-1}(\mathbf{p}) = (uT)^{-1} + \Pi_{\mathcal{G}}(\mathbf{p})$ and $\mathcal{D}_{\mathcal{F}}(\mathbf{p}) = \frac{-\Pi_{\mathcal{F}}(\mathbf{p})\mathcal{D}_{\mathcal{G}}^2(\mathbf{p})}{1-\mathcal{D}_{\mathcal{G}}(\mathbf{p})\Pi_{\mathcal{F}}(\mathbf{p})}$ with

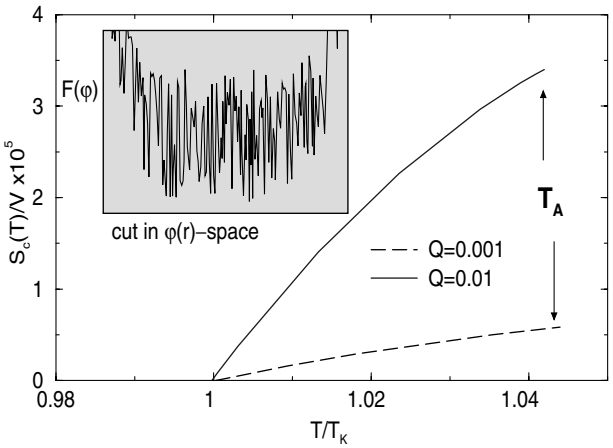

FIG. 1. Configurational entropy density, S_c/V, as a function of T/T_K for $Q = 0.01$ and $r_0 = -10$ ($T_K = 1.7586$) and $Q = 0.001$ and $r_0 = -6$ ($T_K = 1.0422$). In both cases $u = \Lambda = 1$ is used. Note the strong Q dependence of S_c. The inset shows a typical energy landscape for finite S_c.

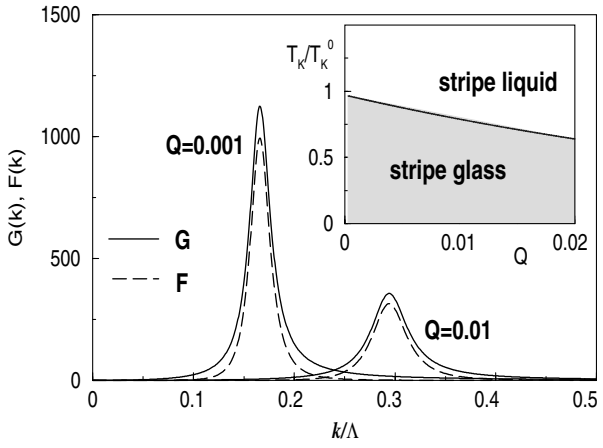

FIG. 2. Momentum dependence of the instantaneous ($\mathcal{G}$) and long-time ($\mathcal{F}$) charge correlation function for the same parameters as in Fig. 1 at $T = T_A$. The inset shows the glass liquid phase diagram as a function of Q for $r_0 = -10$.

polarization functions $\Pi_{\mathcal{A}}(\mathbf{p}) = \int \frac{d^3q}{(2\pi)^3} \mathcal{A}(\mathbf{q}+\mathbf{p})\mathcal{A}(\mathbf{q})$. The set of equations is closed by the Dyson equation: $\mathcal{G}^{-1}(\mathbf{k}) = \mathcal{G}_0^{-1}(\mathbf{k}) + \Sigma_{\mathcal{G}}(\mathbf{k})$ for the diagonal elements, and $\mathcal{F}(\mathbf{k}) = \frac{-\mathcal{G}^2(\mathbf{k})\Sigma_F(\mathbf{k})}{1-\mathcal{G}(\mathbf{k})\Sigma_F(\mathbf{k})}$ for the off-diagonal elements, respectively. $\mathcal{G}_0^{-1}(\mathbf{q}) = r + q^2 + Qq^{-2}$ is the inverse Hartree propagator. Note, all momentum integrations have to be cut off at $|\mathbf{p}| = \Lambda$, which is of the order of an inverse lattice constant. Once the $\mathcal{G}_{ab}$ and $\mathcal{D}_{ab}$ are determined the free energy becomes

$$F(m)/(2mT) = \mathrm{tr}\log\mathcal{G}^{-1} + \mathrm{tr}\log\mathcal{D}^{-1} - \mathrm{tr}\Sigma\mathcal{G}\,. \quad (9)$$

After performing the trace in replica space for arbitrary integer m and analytical continuation to $m \to 1$, S_c follows from Eq. (5). One finds immediately that $S_c = 0$ if $\mathcal{F}(\mathbf{k})$ vanishes. In the following we discuss the numerical solution of this set of coupled integral equations.

In Fig. 1, S_c/V is shown for two different Q values as a function of T. At T_A the long time correlation function $\mathcal{F}(k)$ emerges leading to $S_c > 0$ and a glassy dynamics sets in. The corresponding free energy landscape is schematically illustrated in the inset. Just as in mean field Potts glasses with quenched randomness [27], S_c vanishes at a lower temperature T_K. At T_K the entropy of the amorphous stripe solid equals that of the stripe liquid. There is no entropic advantage anymore to be in a liquid state, leading to an obligatory glass transition no matter how slow the cooling rate. The laboratory glass temperature T_G will lie somewhere between T_K and T_A and cannot be determined within our theory. We also find that T_K and T_A are only weakly decreasing for increasing Q; see inset of Fig. 2. Both temperatures remain finite for $Q \to 0$. However, $S_c(Q \to 0) \to 0$, i.e., the fragility $\propto \frac{dS_c}{dT}$ of the glass vanishes. In other words, the larger the modulation length, the smaller the number of metastable states. Because of the $1/r$ interaction, the limit $Q \to 0$ does not smoothly connect to the behavior at $Q = 0$. If one includes a finite screening length l_s in Eq. (1), we expect that the glassy state disappears for $Q \ll l_s^{-4}$.

In Fig. 2, the instantaneous $[\mathcal{G}(k)]$ and long-time $[\mathcal{F}(k)]$ charge correlation functions are shown at $T = T_A$. Even though no charge ordering occurs, the pronounced peaks at finite k demonstrate that there is a modulated state with strong short range correlations, $l_m < \xi$. Also, since $\mathcal{F}(k) \lesssim \mathcal{G}(k)$, the modulated state exhibits an anomalous dynamics, where long time correlations are only slightly reduced compared to instantaneous correlations. Introducing a Debye-Waller factor, $W = -\log(\mathcal{F}/\mathcal{G})$ for k close to the peak maximum, gives $W = 0.12$ (0.13) for $Q = 0.01$ (0.001). The modulation length (inverse peak positions) is $3.5\Lambda^{-1}$ ($6\Lambda^{-1}$) and the correlation length is $45\Lambda^{-1}$ ($80\Lambda^{-1}$) for $Q = 0.01$ (0.001).

Because of the competing interactions in Eq. (1), an entropy crisis occurs, causing a transition into a glass. This purely thermodynamic characterization of the spectrum of metastable states is only the first important step for understanding glassiness, and the investigation of dynamical features is an even bigger challenge, because it requires going beyond mean field theory. An argument based on "entropic droplets" explains quite well the phenomenology of viscous liquid dynamics [27] and can even be made semiquantitative [31]. Here we apply these arguments to the present stripe model. The entropic droplet argument recognizes an intrinsic instability of the homogeneous metastable solutions, as characterized by Eq. (7): namely, creating a droplet of one metastable solution within another costs free energy that can at most scale as a surface energy but the exponential number of configurations gives an entropic driving force for such a droplet that scales with V. A mosaic structure hence will form. The activation free energy of turning over a single region can be computed where the entropic gain is given by TS_c. A renormalization group

calculation, based on Ref. [32], leads to a size dependent surface tension $\sigma(R) = \sigma_0(R\Lambda)^{-\theta}$ with $\theta = \frac{d-2}{2}$ reflecting the fact that the interface between two states is wetted by intermediate states. This analysis leads to a characteristic energy barrier $\Delta E \propto [TS_c(T)]^{-1}$ which implies a relaxation time obeying a Vogel-Fulcher law [27]

$$\tau \propto \exp\left(\frac{DT_K}{T - T_K}\right). \tag{10}$$

An estimate for the surface tension $\sigma_0 \approx |r_0|/(u\xi)$ for the stripe model yields $R_0^3 \approx \Lambda^{-1}(V\sigma_0/TS_c)^2$ for the droplet volume and $D = 3V\sigma_0^2/(\Lambda T^2 T_K \partial S_c/\partial T|_{T_K})$. Using our numerical results for S_c, this leads to $D \approx 60$–200, typical for moderately fragile and strong glasses, and droplet sizes $R_0 \approx (25$–$50)\Lambda^{-1} \approx (5$–$10)l_m$. Note that this estimate is only qualitative since $R_0 \approx \xi$ and a real separation of scales never occurs. The droplet picture implies that the glass state breaks up into domains of different metastable states, separated by wetted surfaces, built by intermediate states. This physical picture is very similar to the conclusions made in Ref. [10] based on NMR experiments.

In summary, we have shown that an exponentially large number of metastable configurations emerges in a system with competing interactions on different length scales, leading to a glass transition and anomalous long-time dynamics. This glass state is self-generated, implying that the barriers characterizing the activated dynamics are rather universal and should not depend on details like added impurities, but only on the generic interactions on short and long scales, i.e., the magnetic exchange interactions and the Coulomb interaction. Furthermore, we showed that the magnitude of the frustration controls the fragility of the glass transition. Finally, following Ref. [27], we argued that the configurational entropy causes a breakup of the stripe glass into a mosaic of domains or droplets, built up by the various metastable states, allowing us to estimate time scales of motions. This causes an intrinsic inhomogeneity of all relevant correlation functions, modulation, and correlation lengths, etc. in the amorphous glassy state.

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