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A simple theory for the dynamics of mean-field-like models of glass-forming fluids

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We propose a simple theory for the dynamics of model glass-forming fluids, which should be solvable using a mean-field-like approach. The theory is based on transparent physical assumptions, which can be tested in computer simulations. The theory predicts an ergodicity-breaking transition that is identical to the so-called dynamic transition predicted within the replica approach. Thus, it can provide the missing dynamic component of the random first order transition framework. In the large-dimensional limit the theory reproduces the result of a recent exact calculation of Maimbourg *et al.* [PRL **116**, 015902 (2016)]. Our approach provides an alternative, physically motivated derivation of this result.

In the last decade, the static component of the general theoretical framework known as the random first order transition (RFOT) theory [1–3] was developed into a consistent, albeit arguably mean-field-like, description of the glass transition [4–6]. This has been achieved by a combination of the generalization of the replica approach to amorphous systems without quenched disorder [7] with the theoretical apparatus of the liquid state theory [8]. More recently, it has been realized that in the limit of large spatial dimension the mean-field approach becomes exact and the complete theory of the glass (and jamming) transition in the infinite-dimensional hard-sphere system has been worked out [9, 10].

On the other hand, the dynamic component of the RFOT approach has not been advanced to the same degree. This is disappointing since the glass transition, as observed either in a laboratory or a computer experiment, manifests itself most clearly through the enormous slowing down of the dynamics. We note that within the original p -spin version of the RFOT approach [2] (and within the simplified hard-sphere calculation of Ref. [1]) static and dynamic components of the theory were fully consistent. However, there is no finite-dimensional dynamic theory consistent with the advanced version of the replica-based static approach. In contrast, in the limit of large spatial dimension the dynamics of the hard-sphere system has been solved [11]. The many-body problem has been reduced to a one-dimensional stochastic equation with colored noise, which is determined self-consistently. At high density, this equation predicts an ergodicity-breaking transition which is fully consistent with the so-called dynamic transition predicted by the large-dimensional replica calculation. This suggests that it should be possible to come up with dynamical theories that agree with static approaches. Unfortunately, the physical content of the Maimbourg *et al.* result is a bit obscured by a rather long derivation.

One should note at this point that until the formulation of the static replica approach, the mode-coupling theory of the glass transition [12] was the most successful quantitative description of the glass transition and for a long time it was considered to be the dynamic mean-

field theory of this transition (indeed, it was featured in this role in the original RFOT theory papers). However, it was discovered in the last decade [13, 14] that in the large-dimensional limit the ergodicity-breaking transition predicted by the mode-coupling theory is different from the dynamic transition of the replica approach (which becomes exact in the large-dimensional limit). This result showed that the mode-coupling theory does not become exact in the large-dimensional limit and it suggested that this theory is not the proper mean-field theory of the glass transition in finite dimensions. The somewhat uncertain status of the mode-coupling theory [15] is disappointing in view of the fact that it is the starting point for most attempts to go beyond a mean-field-like description of the dynamics [16–18].

Our goal in this Letter is to present a simple theory for glassy dynamics, which predicts ergodicity-breaking transitions consistent with dynamic transitions predicted by the replica approach. This theory can become the missing dynamic component of the RFOT approach. The theory is based on transparent physical assumptions. The most important assumption is that there should be no “loops” in the dynamics (this notion and its consequences are discussed in the following). In principle, this assumption makes the theory applicable only to mean-field-like models of glass-forming fluids. In particular, we show that the large-dimensional limit of our theory coincides with the exact result derived by Maimbourg *et al.* In finite dimensions the situation is a bit more complex. Additional assumptions of the theory, the most prominent one being the Gaussian character of the single-particle motion, make it only an approximate description of the dynamics of finite-dimensional mean-field-like models. On the other hand, analogous assumptions are used in the replica theory description of finite-dimensional mean-field-like models [4] (in principle, one can avoid these assumptions; in practice, it might be easier to do this using the cavity formalism of Mézard *et al.* [19], which is physically equivalent to the replica approach). The result is that the ergodicity-breaking transition predicted by our theory for mean-field-like models in finite dimensions is the same as the dynamic transition

predicted by the replica description.

To make our considerations more specific, we use the Mari-Krzakala-Kurchan model [20]. We consider M particles in d dimensional space, evolving with Brownian dynamics. Any given particle interacts, *via* a spherically symmetric potential $V(r)$, only with N other particles, with $N \ll M$. The network specifying inter-particle interactions forms a quenched random tree-like graph. This graph does not have “loops” and thus dynamical events in which two interacting particles interact with the same third particle are absent. This fact makes the model solvable *via* a mean-field-like approach (at least in principle). In particular, for this model the pair correlation function is equal to the Boltzmann factor, $g(r) = e^{-\beta V(r)}$, with $\beta = 1/T$ (we use a system of units in which $k_B = 1$). At the initial time the particles are distributed according to the canonical ensemble. We note that in the large-dimensional limit the present model system and any system with short-range interactions are identical. This is due to the fact that in large dimensions the probability of “loops” is vanishingly small for geometric reasons.

The first (although not the most important) assumption of our approach is concerned with the description of the single-particle motion: we assume that the motion of one selected (tagged) particle in the fluid of interacting particles can be described by the following generalized Langevin equation with Gaussian noise,

$$\gamma \dot{\mathbf{r}}_1(t) = - \int_0^t M^{\text{irr}}(t-t') \dot{\mathbf{r}}_1(t') dt' + \boldsymbol{\eta}_1(t) + \boldsymbol{\xi}_1(t). \quad (1)$$

Here, γ is the friction coefficient of an isolated particle and $M^{\text{irr}}(t)$ is the irreducible memory function describing the average response of the other particles of the fluid to the motion of the tagged particle. The memory function is essentially a friction kernel, *i.e.* the first term on the right-hand-side of Eq. (1) is the internal friction force experienced by the tagged particle due to the presence of other particles. Next, $\boldsymbol{\eta}_i(t)$ in Eq. (1) is a Gaussian colored noise describing the fluctuating force acting on particle i originating from the presence of the other particles. In equilibrium, the noise should be related to the memory function by a fluctuation-dissipation relation,

$$\langle \boldsymbol{\eta}_i(t) \boldsymbol{\eta}_j(t') \rangle = T \delta_{ij} \mathbf{I} M^{\text{irr}}(t-t'), \quad (2)$$

where \mathbf{I} is the unit tensor. Finally, $\boldsymbol{\xi}_i(t)$ in Eq. (1) is a Gaussian white noise acting on the particle i , with autocorrelation function $\langle \boldsymbol{\xi}_i(t) \boldsymbol{\xi}_j(t') \rangle = 2\gamma T \delta_{ij} \mathbf{I} \delta(t-t')$.

The generalized Langevin equation (1) can be justified as follows. Using the standard projection operator approach one can derive an exact but formal equation for the time evolution of the tagged density auto-correlation function, which involves a wave-vector dependent irreducible memory function. From the evolution equation one can derive an equation of motion for the tagged particle’s mean-square displacement. The latter equation in-

volves the zero-wave-vector limit of the irreducible memory function. If the particle’s motion can be described by a Gaussian stochastic process, then one can deduce Eq. (1) from the time evolution of the tagged particle’s mean-square displacement. It is possible that an exact but formal equation similar to Eq. (1) could be derived, but with some kind of a position dependent irreducible memory function and non-Gaussian noise.

The second, somewhat more technical, assumption of our approach is concerned with the irreducible memory function. One can derive an exact but formal expression for this function as an autocorrelation function of the total force acting on the tagged particle evolving with the so-called irreducible dynamics [21],

$$M^{\text{irr}}(t) = \beta \left\langle \hat{\mathbf{k}} \cdot \mathbf{F}_1 e^{\Omega^{\text{irr}} t} \hat{\mathbf{k}} \cdot \mathbf{F}_1 \right\rangle. \quad (3)$$

Here $\hat{\mathbf{k}}$ is a unit vector, $\mathbf{F}_1 = \sum_{i>1} \mathbf{F}_{1i}$ is the total force acting on the tagged particle (the summation extends over the particles that particle 1 interacts with) and Ω^{irr} is the irreducible evolution operator [21–23] (note that our definition of the memory function includes an additional factor β compared with the definition of Maimbourg *et al.*). Here, following Maimbourg *et al.*, we will assume that the memory function can be obtained from the *pair* force evolving with the *standard* dynamics,

$$\begin{aligned} M^{\text{irr}}(t) &\approx \beta \sum_{i>1} \left\langle \hat{\mathbf{k}} \cdot \mathbf{F}_{1i} e^{\Omega t} \hat{\mathbf{k}} \cdot \mathbf{F}_{1i} \right\rangle \\ &= \beta \sum_{i>1} \left\langle \hat{\mathbf{k}} \cdot \mathbf{F}_{1i}(t) \hat{\mathbf{k}} \cdot \mathbf{F}_{1i}(0) \right\rangle, \end{aligned} \quad (4)$$

where Ω is the standard Smoluchowski operator describing the motion of interacting Brownian particles. Assumption (4) is analogous to the central assumption of the mode-coupling theory, where in the formal expression for the memory function a factorization approximation *and* replacement of the irreducible dynamics by the standard dynamics are applied at the same step [12, 24].

The validity of assumption (4) can be checked almost directly. One could evaluate the right-hand-side of Eq. (4) in a Brownian dynamics computer simulation and then calculate the time-dependence of the mean-square displacement from this approximate irreducible memory function. This calculated approximate mean-square displacement could then be compared to the mean-square displacement measured in the same simulation.

In general, Eq. (4) is only an approximation. For example, for the finite-dimensional model with short-range interactions the initial ($t = 0$) values of the exact memory function (3) and the approximation (4) are different. However, for a Mari-Krzakala-Kurchan model, due to the loop-less structure of the inter-particle interactions network, the initial values of these functions are the same.

The second assumption, Eq. (4), suggests that one can evaluate the memory function by calculating the force

between two selected particles, evolving with the standard dynamics, given that at the initial time these two particles were distributed according to probability distribution $\propto \hat{\mathbf{k}} \cdot \mathbf{F}_{12}g(r_{12})$, where $g(r_{12})$ is the equilibrium pair correlation function.

Let us now consider the two-particle dynamics. The force acting on one of these particles consists of the following parts. The first part is the force due to the second particle. The second part describes the interaction with the other particles of the fluid, *i.e.* particles different from the interacting pair of particles. The latter force has the structure similar to the force acting on one particle, Eq. (1). It consists of an average part and a fluctuating part. For a Mari-Krzakala-Kurchan model (and for any model in the large-dimensional limit) the force acting on the first particle and originating from other particles of the fluid should be independent of the state of the second particle (and *vice versa*). This is due to the fact that in these models the particles that interact with the first particle (and the particles that interact with these particles *etc.*) do not interact with the second particle. We have to emphasize here that the argument described in this paragraph relies on our *most important* assumption that the structure of the inter-particle interactions is such that “loops” are absent both in the statics and in the dynamics.

The argument formulated above leads us to assume the following equations of motion for the dynamics of the interacting pair of particles,

$$\gamma \dot{\mathbf{r}}_1(t) = \mathbf{F}(\mathbf{r}_{12}(t)) \quad (5)$$

$$- \int_0^t M^{\text{irr}}(t-t') \dot{\mathbf{r}}_1(t') dt' + \boldsymbol{\eta}_1(t) + \boldsymbol{\xi}_1(t)$$

$$\gamma \dot{\mathbf{r}}_2(t) = -\mathbf{F}(\mathbf{r}_{12}(t)) \quad (6)$$

$$- \int_0^t M^{\text{irr}}(t-t') \dot{\mathbf{r}}_2(t') dt' + \boldsymbol{\eta}_2(t) + \boldsymbol{\xi}_2(t).$$

To calculate the inter-particle force we only need the relative position, $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$. The equation of motion for the relative position can be obtained from Eqs. (5-6),

$$\tilde{\gamma} \dot{\mathbf{r}}(t) = \mathbf{F}(\mathbf{r}(t)) - \int_0^t \tilde{M}(t-t') \dot{\mathbf{r}}(t') dt' + \boldsymbol{\eta}(t) + \boldsymbol{\xi}(t) \quad (7)$$

where $\tilde{\gamma} = \frac{1}{2}\gamma$, $\tilde{M}(t) = \frac{1}{2}M^{\text{irr}}(t)$, $\boldsymbol{\eta}(t) = \frac{1}{2}(\boldsymbol{\eta}_1(t) - \boldsymbol{\eta}_2(t))$, $\boldsymbol{\xi}(t) = \frac{1}{2}(\boldsymbol{\xi}_1(t) - \boldsymbol{\xi}_2(t))$ and thus $\langle \boldsymbol{\xi}(t) \boldsymbol{\xi}(t') \rangle = 2\gamma T \mathbf{I} \delta(t-t')$ and $\langle \boldsymbol{\eta}(t) \boldsymbol{\eta}(t') \rangle = \mathbf{I} T \tilde{M}(t-t')$. We note that Eq. (7) is to be solved with the initial condition distributed according to $P(\mathbf{r}) = n \hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r}) g(r)$ where $n = N/V$ is the number density of particles interacting with the tagged particle and $g(r) = e^{-\beta V(r)}$ is the pair correlation function for the Mari-Krzakala-Kurchan model. Then, the memory function is given by

$$\tilde{M}(t) = \frac{1}{2} M^{\text{irr}}(t) = \frac{n\beta}{2} \int d\mathbf{r} \langle \hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r}(t)) \hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r}) \rangle g(r), \quad (8)$$

where $\mathbf{r}(t=0) = \mathbf{r}$ and the averaging is over noises in Eq. (7).

We note that even though the noise $\boldsymbol{\eta}(t)$ is Gaussian (since it is defined as a difference of two Gaussian noises), the process describing the pair dynamics, *i.e.* $\mathbf{r}(t)$, is not Gaussian. This is due to the fact that the relation between these two processes, *i.e.* Eq. (7), is non-linear.

In principle, the self-consistent solution of Eq. (7) determines the irreducible memory function. At present, an analytic solution of this equation is not available. We can, however, show that at high enough density Eqs. (7-8) predict an ergodicity-breaking transition. To this end we can use the argument similar to that used by Maimbourg *et al.* (see Sec. V.A.1 of Supplemental Material for Ref. [11]).

We assume that at high enough density or low enough temperature a two-step relaxation process sets up, with both the mean-square displacement and the irreducible memory function being temporarily arrested around their respective plateau values. For times in the plateau region, both parts of the noise $\boldsymbol{\eta}(t)$ and the corresponding part of the memory function can be treated as adiabatically slow. It can be showed (see Supplemental Material) that the plateau value of the memory function, \tilde{M}_{EA} , can be expressed as follows,

$$\tilde{M}_{\text{EA}} = \frac{n\beta}{2} \int d\mathbf{s} P_{\text{slow}}(\mathbf{s}) \langle \hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r}) \rangle_{\mathbf{s}}^2 \quad (9)$$

where the distribution of the slow variable \mathbf{s} (which is a linear combination of the slow part of the noise and the initial condition) reads

$$P_{\text{slow}}(\mathbf{s}) = \int \frac{d\mathbf{r}}{(2\pi T \tilde{M}_{\text{EA}})^{d/2}} e^{-\beta V(\mathbf{r}) - \frac{(\mathbf{s} - \tilde{M}_{\text{EA}} \mathbf{r})^2}{2T \tilde{M}_{\text{EA}}}} \quad (10)$$

and the conditional average for a given value of the slow variable is defined as

$$\langle f(\mathbf{r}) \rangle_{\mathbf{s}} = \frac{\int d\mathbf{r} e^{-\beta(V(\mathbf{r}) + \tilde{M}_{\text{EA}} \mathbf{r}^2/2 - \mathbf{s} \cdot \mathbf{r})} f(\mathbf{r})}{\int d\mathbf{r} e^{-\beta(V(\mathbf{r}) + \tilde{M}_{\text{EA}} \mathbf{r}^2/2 - \mathbf{s} \cdot \mathbf{r})}}. \quad (11)$$

We should note that the specific form of Eqs. (10-11) follows from the assumed Gaussian character of the noise.

Eq. (9) is a self-consistent equation for the plateau value of the memory function, \tilde{M}_{EA} . A non-zero solution of this equation signals an ergodicity-breaking transition. It can be showed (see Supplemental Material) that Eq. (9) is equivalent to the equation determining the dynamic transition within the replica approach [25]. One should recall that the latter equation was derived using an assumption of a Gaussian “cage”, which corresponds to our assumption of the Gaussian character of the single-particle motion and of the noise. The ergodicity-breaking transition predicted by Eq. (9) takes place below a crossover density determined by power-law fits to the computer simulation data, with the difference

between the two decreasing with increasing dimension [25].

Now, let us show that in the large-dimensional limit our theory is identical to the exact result of Maimbourg *et al.* First, we use the fact that in the large-dimensional limit the relative motion of two particles that are interacting at the initial time (which is the process described by Eq. (7) with the initial condition distributed according to $P(\mathbf{r}) = n\hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r})g(r)$) proceeds predominantly along the original direction of the relative coordinate, *i.e.* along $\mathbf{r}(t=0)$. In other words, in the large-dimensional limit $\partial_t \hat{\mathbf{r}}(t)$ can be neglected. This allows us to focus on the equation of motion for the inter-particle distance $r(t) \equiv |\mathbf{r}(t)|$. Using Itô's convention we obtain,

$$\begin{aligned} \tilde{\gamma} \dot{r}(t) = & F(r(t)) - \hat{\mathbf{r}}(t) \cdot \int_0^t \tilde{M}(t-t') \dot{\mathbf{r}}(t') dt' \\ & + T \frac{d-1}{r(t)} + \hat{\mathbf{r}}(t) \cdot \boldsymbol{\eta}(t) + \hat{\mathbf{r}}(t) \cdot \boldsymbol{\xi}(t), \end{aligned} \quad (12)$$

where $F(r(t)) = \hat{\mathbf{r}}(t) \cdot \mathbf{F}(\mathbf{r}(t))$ and the first term in the second line originates from Itô's lemma. Since $\hat{\mathbf{r}}(t) \approx \hat{\mathbf{r}}(t')$, we can express the second term on the right-hand-side of Eq. (12) in terms of $r(t')$. Next, we use the scaling relationships introduced by Maimbourg *et al.*, which can also be deduced from the large-dimensional limit of Eq. (9): $r(t) = \sigma(1 + h(t)/d)$, $\hat{\gamma} = \tilde{\gamma}\sigma^2/d^2 = \gamma\sigma^2/(2d^2)$, $F(h) = \frac{\sigma}{d}F(r)$ and $M(t) = \frac{\sigma^2}{d^2}\tilde{M}(t)$, with σ being the particle diameter. Then, in the large-dimensional limit we get from Eq. (12) the following one dimensional stochastic equation for the “gap” $h(t)$,

$$\dot{h}(t) = -w'(h(t)) + \int_0^t M(t-t') \dot{h}(t') dt' + \eta(t) + \xi(t), \quad (13)$$

where the effective force is given by $-w'(h) = F(h) + T$ and the noises η and ξ satisfy fluctuation-dissipation relations $\langle \eta(t)\eta(t') \rangle = TM(t-t')$ and $\langle \xi(t)\xi(t') \rangle = 2\gamma T\delta(t-t')$. Finally, from the memory function expression (4) we can get an expression for the re-scaled function $M(t)$,

$$M(t) = \frac{\beta\hat{\phi}}{2} \int dh \langle F(h)F(h(t)) \rangle e^{-\beta w(h)}, \quad (14)$$

where $h(t=0) = h$, $\hat{\phi}$ is the re-scaled volume fraction, $\hat{\phi} = n\mathcal{V}_d 2^d/d$, with \mathcal{V}_d being the volume of a d -dimensional sphere of radius $\sigma/2$, and the averaging is over noises in Eq. (13).

Eqs. (13-14) are identical to the equations derived by Maimbourg *et al.* In particular, these equations predict an ergodicity-breaking transition at $\hat{\phi}_d = 4.807$.

Let us comment on the connection of the present approach and the mode-coupling theory. The most important approximation of the latter theory is the factorization approximation in which a four-point correlation

function is replaced by a product of two-point functions. In the present approach, the analogue of this approximation would be neglecting the force term in the stochastic equation of motion for the relative position, Eq. (7). However, if one were just to neglect the force term in Eq. (7), the expression (8) for the memory function would give an infinite result. The way out is to incorporate one of the additional approximations of the mode-coupling theory [12] and to combine discarding the force term with replacing the “bare” forces in the memory function expression (8) by renormalized forces given by the derivatives of the direct correlation function. In the present case of the Mari-Krzakala-Kurchan model the last step amounts to the replacement $\mathbf{F}(\mathbf{r}) \rightarrow T\partial_{\mathbf{r}}e^{-\beta V(r)}$ in expression (8). It can be showed that this procedure results in the following self-consistent equation for the plateau value of the memory function, $M_{\text{EA}}^{\text{mct}}$, where the superscript mct indicates a mode-coupling-like approximation,

$$\begin{aligned} \tilde{M}_{\text{EA}}^{\text{mct}} = & \frac{n\beta}{2} \int \frac{d\mathbf{r}d\mathbf{r}'}{(4\pi T/\tilde{M}_{\text{EA}}^{\text{mct}})^{d/2}} e^{-\frac{(\mathbf{r}-\mathbf{r}')^2 \tilde{M}_{\text{EA}}^{\text{mct}}}{4T}} \\ & \times g(r)\hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r})g(r')\hat{\mathbf{k}} \cdot \mathbf{F}(\mathbf{r}'). \end{aligned} \quad (15)$$

By rewriting the right-hand-side of this equation as an integral in reciprocal space we can show that it is identical to Eq. (4.6') of Ref. [1] (with additional factors of 2 in a couple of places). In the large-dimensional limit Eq. (15) predicts an ergodicity-breaking transition at $\hat{\phi}_d^{\text{mct}} = \sqrt{8\pi e} = 8.265$. This finding agrees perfectly with the numerical result of Ikeda and Miyazaki [14] obtained by taking the large-dimensional limit of the standard mode-coupling equation for the non-ergodicity parameter and assuming a Gaussian form of this parameter. We note that the mode-coupling-like version of our theory predicts the ergodicity-breaking transition at a higher value of the volume fraction. This is reasonable in that the mode-coupling-like approach neglects direct interaction between the two particles; replacing “bare” forces by renormalized ones only partially compensates for this.

In summary, we presented here a simple theory for the dynamics of models of structural glasses which should be solvable using a mean-field-like approach. The ergodicity-breaking transition predicted by our theory coincides with the dynamic transition of the replica approach. Thus, our theory provides the dynamic counterpart of the static replica approach.

In our theory, any given particle interacts explicitly only with one other particle of the system. Other interactions are accounted for by a combination of a friction force and a fluctuating force.

In the large-dimensional limit our theory reduces itself to the result of the exact calculation of Maimbourg *et al.* and thus it provides an alternative, physically motivated derivation of this result. In finite dimensions it suffers from the same problem as the current version of

the replica approach, *i.e.* from an additional assumption of the Gaussian character of the single-particle dynamics. It would be very interesting to develop an analogue of a cavity approach of Mézard *et al.* which could possibly overcome this problem.

The simplest generalization of our theory to standard finite-dimensional systems with non-trivial local structure is to replace the true potential in Eqs. (7-8) with the potential of the mean force $V^{\text{mf}}(r) = -T \ln g(r)$. It can be showed (using the methods presented in the Supplemental Material) that with this modification our theory predicts an ergodicity-breaking transition which coincides with the dynamic transition obtained within a recent version of the replica theory for a standard finite-dimensional system [26]. Since the result of the latter approach agrees quite well with computer simulations, this suggests that our theory could be generalized to describe standard finite-dimensional systems. At the same time, our theory can shed some light onto somewhat abstract considerations of the replica approach. For example, the result of the above described modification suggests that the replica approach, at least as far as location of the dynamic transition is concerned, neglects “loops” and thus approximates the standard hard-sphere system by a Mari-Krzakala-Kurchan system with particles interacting via the potential of mean force.

Finally, it would be interesting to start from equations of motion for density fields and to develop an approximate theory by keeping only dynamical events which are included in the present approach. This might be another avenue that would allow us to avoid or relax the assumption of Gaussian fluctuations and thus arrive at a theory that both provides a reasonable, albeit mean-field-like, description of the dynamics of finite-dimensional systems and has the correct large-dimensional limit.

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