Supplementary Information for

Evolved interactions stabilize many coexisting phases in multicomponent liquids

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Other supplementary materials for this manuscript include the following:

> Movie S1

Contents

1. Derivation of the simple dynamical system to detect equilibrium states

We here give a detailed derivation of the simple dynamical system that we use in the main text to determine equilibrium states of the multicomponent liquid. In particular, we demonstrate below that the stationary states of this dynamical system correspond to actual equilibrium solutions.

A. Equilibrium conditions of the physical system. The conditions for an equilibrium between two phases n and m read

$$\hat{\mu}_i^{(n)} = \hat{\mu}_i^{(m)} \qquad\qquad \text{and} \qquad\qquad \hat{P}^{(n)} = \hat{P}^{(m)} \qquad\qquad \forall\, n, m, i\,, \qquad\qquad [\text{S1}]$$

where i enumerates the N components; see Eq. 3 in the main text. The non-dimensional chemical potentials and pressures are

$$\hat{\mu}_i^{(n)} = \ln(\phi_i^{(n)}) - \ln(\phi_0^{(n)}) + \sum_{j=1}^{N} \chi_{ij}\phi_j^{(n)} \qquad\qquad \hat{P}^{(n)} = -\ln(\phi_0^{(n)}) + \sum_{i,j=1}^{N} \frac{\chi_{ij}}{2}\phi_i^{(n)}\phi_j^{(n)}\,. \qquad\qquad [\text{S2}]$$

In general, the multiphase system is specified by the phase volumes $V^{(n)}$ together with either all particle numbers, $N_i^{(n)}$, or all volume fractions $\phi_i^{(n)} = \nu N_i^{(n)}/V^{(n)}$. Here, $V^{(n)}$ and $N_i^{(n)}$ are extensive quantities that grow with system size, while the fractions $\phi_i^{(n)}$ are the intensive quantities that appear in the Eqs. **S1**–**S2**.

B. Dynamics of the full physical system. To determine the equilibrium states satisfying Eqs. **S1**, we first discuss the dynamics of the physical system, specified by the intensive quantities $\phi_i^{(n)}$ together with the extensive volumes $V^{(n)}$ of all phases. We can then express the rate of change of the free energy $F = \sum_{n=1}^{M} V^{(n)} f(\phi_i^{(n)})$ as

$$\partial_t F = -\sum_{n=1}^{M} P^{(n)} \partial_t V^{(n)} + \sum_{i=1}^{N}\sum_{n=1}^{M} \mu_i^{(n)} \partial_t N_i^{(n)}\,. \qquad\qquad [\text{S3}]$$

Here, we consider the spontaneous relaxation to equilibrium, implying dynamics that decrease F continuously, $\partial_t F \leq 0$. Using linear non-equilibrium thermodynamics (1), one can show that the exchange of volume is driven by pressure differences and differences in chemical potential imply changes in particle numbers,

$$\partial_t V^{(n)} = \nu \sum_{m} \alpha^{(nm)}(\hat{P}^{(n)} - \hat{P}^{(m)}) \qquad\qquad \partial_t N_i^{(n)} = \sum_{m} \beta_i^{(nm)}(\hat{\mu}_i^{(m)} - \hat{\mu}_i^{(n)})\,, \qquad\qquad [\text{S4}]$$

where the kinetic coefficients $\alpha^{(nm)}$ and $\beta_i^{(nm)}$ need to be symmetric, $\alpha^{(nm)} = \alpha^{(mn)}$ and $\beta_i^{(nm)} = \beta_i^{(mn)}$. We can use this together with Eq. **S3** to show explicitly that the free energy cannot increase under these conditions,

$$\frac{\partial_t F}{k_\mathrm{B}T} = -\sum_{n,m=1}^{M} \frac{\alpha^{(nm)}}{2}\left(\hat{P}^{(n)} - \hat{P}^{(m)}\right)^2 - \sum_{i=1}^{N}\sum_{n,m=1}^{M} \frac{\beta_i^{(nm)}}{2}\left(\hat{\mu}_i^{(m)} - \hat{\mu}_i^{(n)}\right)^2\,. \qquad\qquad [\text{S5}]$$

This implies that the following three statements about the system's state are all equivalent: (i) F is at a stationary point, $\partial_t F = 0$; (ii) The dynamics given in Eq. **S4** are at a stationary point; (iii) The system fulfills the equilibrium conditions Eq. **S1** and the volume and particle number constraints. We can thus determine solutions to Eq. **S1** using the dynamics given in Eq. **S4** to relax initial conditions to a stationary state.

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A concrete implementation of the relaxation dynamics requires sensible choices for the kinetic coefficients. We use $\alpha^{(nm)} = k$ and $\beta_i^{(nm)} = k\phi_i^{(n)}\phi_i^{(m)}$, where we introduced the relaxation rate k, which defines the non-dimensional time $\hat{t} = kt$. Using $\partial_t \phi_i^{(n)} = [\nu\partial_t N_i^{(n)} - \phi_i^{(n)}\partial_t V^{(n)}]/V^{(n)}$, the dynamics of the system read

$$\partial_{\hat{t}} V^{(n)} = \nu \sum_{m=1}^{M} \left[\hat{P}^{(n)} - \hat{P}^{(m)}\right] \tag{S6a}$$

$$\partial_{\hat{t}} \phi_i^{(n)} = \frac{\nu\phi_i^{(n)}}{V^{(n)}} \sum_{m=1}^{M} \left[\phi_i^{(m)}\left(\hat{\mu}_i^{(m)} - \hat{\mu}_i^{(n)}\right) + \left(\hat{P}^{(m)} - \hat{P}^{(n)}\right)\right] . \tag{S6b}$$

These equations define an initial value problem, which relaxes an initial configuration, $V^{(n)}$ and $\phi_i^{(n)}$, toward equilibrium.

C. Simplified dynamical system. The physical dynamics defined by Eq. **S6** involve both the intensive fractions $\phi_i^{(n)}$ and the extensive volumes $V^{(n)}$. However, the equilibrium conditions **S1** are local statements about the coexistence of phases and thus do not involve extensive quantities. To solve only the coexistence problem, we now seek a dynamical system that only involves intensive variables. Inspecting Eq. **S6b**, we see that the volumes $V^{(n)}$ only affect the rate at which the fractions $\phi_i^{(n)}$ change but not the direction of change. In particular, the nullclines of the dynamics remain invariant. Consequently, we obtain qualitatively similar dynamics by removing the pre-factor,

$$\partial_{\hat{t}} \phi_i^{(n)} = \phi_i^{(n)} \sum_{m=1}^{M} \left[\phi_i^{(m)}\left(\hat{\mu}_i^{(m)} - \hat{\mu}_i^{(n)}\right) + \hat{P}^{(m)} - \hat{P}^{(n)}\right] , \tag{S7}$$

which is identical to the simplified dynamical system given by Eq. 4 in the main text. Fig. S1 provides numerical evidence that these dynamics discover the structure of the phase diagram. However, to be confident that stationary states of the dynamical system correspond to equilibrium states, we have to check the implication in both directions. Clearly, equilibrium states, which fulfill Eq. **S1**, are stationary states of Eq. **S7**. We next demonstrate that the NM stationary state conditions following from Eq. **S7** also imply the equilibrium conditions Eq. **S1**. To do this, we introduce the deviations

$$x_i^{(n)} = \hat{\mu}_i^{(n)} - \bar{\mu}_i \qquad\qquad\qquad y^{(n)} = \hat{P}^{(n)} - \bar{P} \tag{S8}$$

from the means $\bar{\phi}_i = M^{-1}\sum_n \phi_i^{(n)}$, $\bar{\mu}_i = M^{-1}\sum_n \hat{\mu}_i^{(n)}$, and $\bar{P} = M^{-1}\sum_n \hat{P}^{(n)}$. Using this, we can give the equilibrium conditions as $(N+1)M$ equations,

$$x_i^{(n)} = 0 \qquad\qquad\qquad y^{(n)} = 0 \qquad\qquad \forall\, n, i\,, \tag{S9}$$

with $N+1$ constraints, $\sum_n x_i^{(n)} = \sum_n y^{(n)} = 0$, resulting in $(N+1)(M-1)$ independent conditions. In contrast, we have NM conditions for the stationary state,

$$0 = \frac{1}{M}\sum_{m=1}^{M} \phi_i^{(m)} x_i^{(m)} - \bar{\phi}_i x_i^{(n)} - y^{(n)} \qquad\qquad \forall\, n, i\,. \tag{S10}$$

Summing Eq. **S10** over n, we find $0 = \sum_m \phi_i^{(m)} x_i^{(m)}$, which implies

$$0 = \bar{\phi}_i x_i^{(n)} + y^{(n)} \qquad\qquad \forall\, n, i\,. \tag{S11}$$

Note that these are only $N(M-1)$ independent equations since $\sum_n x_i^{(n)} = \sum_n y^{(n)} = 0$. There are N additional equations that follow from Eq. **S10**, resulting in NM independent equations describing the stationary state,

$$0 = \bar{\phi}_i x_i^{(n)} + y^{(n)} \quad \forall\, i \le N, n \le M-1 \qquad\qquad 0 = \sum_{n=1}^{M} \phi_i^{(n)} x_i^{(n)} \quad \forall\, i\,. \tag{S12}$$

To show that these are equivalent to the equilibrium conditions **S9**, we express them as a linear system,

$$0 = \mathcal{A}.X\,, \tag{S13}$$

where $X = (x_1^{(1)}, \ldots, x_N^{(1)}, x_1^{(2)}, \ldots, x_N^{(2)}, \ldots, x_1^{(M-1)}, \ldots, x_N^{(M-1)}, y^{(1)}, y^{(2)}, \ldots y^{(M-1)})$ has $(M-1)(N+1)$ entries, and

$$
\mathcal{A} = \begin{pmatrix}
\begin{matrix} \bar{\phi}_1 & 0 & \cdots & 0 \\ 0 & \bar{\phi}_2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \bar{\phi}_N \end{matrix} & \mathbf{0} & \cdots & \mathbf{0} & \begin{matrix} 1 & 0 & \cdots & 0 \\ 1 & 0 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 1 & 0 & \cdots & 0 \end{matrix} \\
\mathbf{0} & \begin{matrix} \bar{\phi}_1 & 0 & \cdots & 0 \\ 0 & \bar{\phi}_2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \bar{\phi}_N \end{matrix} & \cdots & \mathbf{0} & \begin{matrix} 0 & 1 & \cdots & 0 \\ 0 & 1 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 1 & \cdots & 0 \end{matrix} \\
\vdots & \vdots & \ddots & \vdots & \vdots \\
\mathbf{0} & \mathbf{0} & \cdots & \begin{matrix} \bar{\phi}_1 & 0 & \cdots & 0 \\ 0 & \bar{\phi}_2 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \bar{\phi}_N \end{matrix} & \begin{matrix} 0 & 0 & \cdots & 1 \\ 0 & 0 & \cdots & 1 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & 1 \end{matrix} \\
\begin{matrix} \phi_1^{(1)} & 0 & \cdots & 0 \\ 0 & \phi_2^{(1)} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \phi_N^{(1)} \end{matrix} & \begin{matrix} \phi_1^{(2)} & 0 & \cdots & 0 \\ 0 & \phi_2^{(2)} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \phi_N^{(2)} \end{matrix} & \cdots & \begin{matrix} \phi_1^{(M-1)} & 0 & \cdots & 0 \\ 0 & \phi_2^{(M-1)} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \phi_N^{(M-1)} \end{matrix} & \mathbf{0}
\end{pmatrix}
\tag{S14}
$$

is a matrix of $M \times M$ blocks where each block has the dimension $N \times N$, except in the last column, where the blocks have dimension $N \times (M-1)$. The block size can thus be summarized as

$$
\begin{pmatrix}
N \times N & \cdots & N \times N & N \times (M-1) \\
\vdots & \ddots & \vdots & \vdots \\
N \times N & \cdots & N \times N & N \times (M-1)
\end{pmatrix}.
\tag{S15}
$$

Clearly, this matrix has MN rows and $(M-1)(N+1)$ columns, so it is a square matrix if and only if $M = N+1$.

To show that the stationary state conditions given in Eq. **S10** imply the equilibrium conditions Eq. **S9**, we need to show that the linear system given in Eq. **S13** only has the trivial solution $X = 0$. If $\mathcal{A}$ is a square matrix, this amounts to showing that its determinant is non-zero. We will show below that the non-square case can be treated by investigating the largest square sub matrix $\mathcal{A}_{\mathrm{sq}}$, which is built by dropping the last rows or columns. Defining the relevant dimension $d = \min(M-1, N)$, the determinant of this matrix reads

$$
\det(\mathcal{A}_{\mathrm{sq}}) = (-1)^d \det(\mathcal{P}) \prod_{i=1}^{d} \bar{\phi}_i^{M-2} \prod_{j=d+1}^{N} \bar{\phi}_j^{M-1} ,
\tag{S16}
$$

where we defined the square composition matrix

$$
\mathcal{P} = \begin{pmatrix}
\phi_1^{(1)} & \phi_1^{(2)} & \cdots & \phi_1^{(d)} \\
\phi_2^{(1)} & \phi_2^{(2)} & \cdots & \phi_2^{(d)} \\
\vdots & \vdots & \ddots & \vdots \\
\phi_d^{(1)} & \phi_d^{(2)} & \cdots & \phi_d^{(d)}
\end{pmatrix}.
\tag{S17}
$$

Note that the determinant of $\mathcal{A}_{\mathrm{sq}}$ only vanishes if $\det(\mathcal{P}) = 0$, since $\bar{\phi}_i > 0$. In the following, we analyze the solution space for the three relevant dimensional cases.

C.1. Balanced case of a square matrix. In the balance case, $M = N+1 = d+1$, we have $\mathcal{A} = \mathcal{A}_{\mathrm{sq}}$ and $\mathcal{P}$ describes the full composition. The linear system $0 = \mathcal{A}.X$ could have non-trivial solutions if the determinant of $\mathcal{P}$ vanishes. Generally, $\det(\mathcal{P})$ vanishes if rows or columns are linearly dependent. This is for instance the case when two phases have identical composition, $\phi_i^{(n)} = \phi_i^{(m)}$ for two phases $n \neq m$ and all components i. However, in this case the conditions $\hat{\mu}_i^{(n)} = \hat{\mu}_i^{(m)}$ and $\hat{P}^{(n)} = \hat{P}^{(m)}$ are trivially fulfilled, so we can always ignore identical phases and instead focus on phases with distinct composition. The determinant also vanishes when two species have identical composition in all phases, $\phi_i^{(n)} = \phi_j^{(n)}$ for some $i \neq j$ and all n. This is only possible if they behave identically, $\chi_{ik} = \chi_{jk}$ for all k, in which case we again find $\hat{\mu}_i^{(n)} = \hat{\mu}_j^{(n)}$ and $\hat{P}^{(n)} = \hat{P}^{(m)}$. In this case, the two components can basically be treated as one and thus also ignored. Generally, the determinant also vanishes when a row (column) can be written as a linear sum of the other rows (columns). While we could not identify a mathematical statement that this is impossible, we never observed such a case and will thus not discuss it further. Taken together, we conclude that the stationary state conditions Eq. **S10** imply the equilibrium conditions Eq. **S9** if $M = N+1$.

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C.2. Overdetermined case of few phases. In this case, $\mathcal{A}$ has more rows than columns, so there are more stationary state conditions than equilibrium conditions. This implies $NM > (N+1)(M-1)$, $M < N+1$, and $d = M - 1$. By solving the square sub-problem involving $\mathcal{A}_{\mathrm{sq}}$, we find $x_i^{(n)} = y^{(n)} = 0$. In doing so, we ignored the extra conditions

$$0 = \sum_{n=1}^{M} \phi_i^{(n)} x_i^{(n)} \qquad \text{for} \quad i = M, \dots, N \,, \tag{S18}$$

but these are trivially fulfilled. Taken together, this shows that the overdetermined case implies $x_i^{(n)} = y^{(n)} = 0$.

C.3. Underdetermined case of too many phases. In this case, $\mathcal{A}$ has more columns than rows, $NM < (N+1)(M-1)$, $M > N+1$, and $d = N$. This implies that there are not sufficient stationary state conditions to immediately conclude that the equilibrium conditions hold. However, the variables $x_i^{(n)}$ and $y^{(n)}$ are not truly independent since they are both functions of the composition $\phi_i^{(n)}$; see Eq. **S2**. Combining these conditions, we find

$$\sum_{i=1}^{N} \phi_i^{(n)} \hat{\mu}_i^{(n)} - 2\hat{P}^{(n)} = \sum_{i=1}^{N} \phi_i^{(n)} \ln \phi_i^{(n)} + (1 + \phi_0^{(n)}) \ln \phi_0^{(n)} \qquad \forall n \,, \tag{S19}$$

demonstrating a linear relationship between $\hat{\mu}_i^{(n)}$ and $\hat{P}^{(n)}$ that depends non-linearly on $\phi_i^{(n)}$. Since these conditions must also hold, it is plausible that only NM of the $(N+1)(M-1)$ unknowns of the linear system **S13** are independent. Assuming this is the case, we can solve the linear subsystem of NM dimensions, whose determinant is given in Eq. **S2**. Similarly to the cases discussed above, this system has only the trivial solutions, implying that $x_i^{(n)} = y^{(n)} = 0$ for the first NM unknowns. This shows that most equilibrium conditions follow from the stationary state conditions in the underdetermined case and suggests that the stationary state system does not possess any solutions that violate the equilibrium conditions. Indeed, we never observed any numerical solutions of the stationary state conditions **S10** that violated the equilibrium conditions **S9**.

Note that the underdetermined system is related to Gibbs' phase rule, which implies that at most $N + 1$ phases can have different composition (2). Consequently, the number of undetermined variables we encountered here is exactly the minimal number of phases that needs to have a composition equivalent to other phases. The additional constraints introduced by the fact that $x_i^{(n)}$ and $y^{(n)}$ all depend on $\phi_i^{(n)}$ thus reflect the thermodynamic stability discussed by Gibbs.

2. Distribution of random initial composition

We briefly describe how we choose an initial composition $\vec{\phi} = (\phi_1, \dots, \phi_N)$ for a phase such that all admissible composition vector $(\phi_0, \phi_1, \dots, \phi_N)$, including the solvent fraction ϕ_0, exhibit a uniform distribution. Here, we obviously use $\phi_0 = 1 - \sum_{i=1}^{N} \phi_i$ and we ensure $\phi_i \geq 0$ for $i = 0, 1, \dots, N$. The condition that all fractions sum to one implies correlations between the fractions, but since the geometry of allowed fractions is a simplex in an N-dimensional space, we can determine the marginal distributions along each dimension and choose the fractions iteratively. The associated conditional probabilities are given by

$$P_1(\phi_1) = P(\phi_1; 1, N) \tag{S20a}$$

$$P_2(\phi_2|\phi_1) = P(\phi_2; 1 - \phi_1, N - 1) \tag{S20b}$$

$$\vdots$$

$$P_i(\phi_i|\phi_1, \dots, \phi_{i-1}) = P\left(\phi_i; 1 - \sum_{j=1}^{i-1} \phi_j, N - i + 1\right) \tag{S20c}$$

$$\vdots$$

$$P_N(\phi_N|\phi_1, \dots, \phi_{N-1}) = \frac{1}{1 - \sum_{j=1}^{N-1} \phi_j} \,, \tag{S20d}$$

where the probability density function

$$P(\phi; f, n) = n \, (f - \phi)^{n-1} f^{-n} \tag{S21}$$

describes a particular, scaled Beta distribution defined for $\phi \in [0, f]$. The conditional probabilities given in Eqs. **S20** allow us to draw random variates of $\vec{\phi}$, since we can sample from the one-dimensional distributions for $i = 1, \dots, N$ one after another. Note that the joint probability distribution reads

$$P(\phi_1, \dots, \phi_N) = \prod_{i=1}^{N} P_i(\phi_i|\phi_1, \dots, \phi_{i-1}) = N! \tag{S22}$$

and is thus constant, demonstrating that this is truly a uniform distribution.

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To demonstrate the procedure, we consider the example $N = 2$, where we have $P_1(\phi_1) = 2(1-\phi_1)$ and $P_2(\phi_2|\phi_1) = (1-\phi_1)^{-1}$, implying $P(\phi_1, \phi_2) = P_1(\phi_1)P_2(\phi_2|\phi_1) = 2$. The expectation values are

$$\langle \phi_0 \rangle = \int_0^1 \mathrm{d}\phi_1 \int_0^{1-\phi_1} \mathrm{d}\phi_2 \, P_{12}(\phi_1, \phi_2)\,(1 - \phi_1 - \phi_2) = \frac{1}{3} \tag{S23a}$$

$$\langle \phi_1 \rangle = \int_0^1 \mathrm{d}\phi_1 \int_0^{1-\phi_1} \mathrm{d}\phi_2 \, P_{12}(\phi_1, \phi_2)\,\phi_1 = \frac{1}{3} \tag{S23b}$$

$$\langle \phi_2 \rangle = \int_0^1 \mathrm{d}\phi_1 \int_0^{1-\phi_1} \mathrm{d}\phi_2 \, P_{12}(\phi_1, \phi_2)\,\phi_2 = \frac{1}{3} \tag{S23c}$$

demonstrating that indeed all three components have identical average fraction.

The method presented in the main text relies on initializing the fractions of N components in M different phases to determine coexisting fractions. This takes place by exchanging fractions between the phases until the equilibrium conditions are fulfilled; see detailed discussion in Section 1. Fig. S1 shows that broad initial distributions of the fractions $\phi_i^{(n)}$ typically converge to fewer equilibrium points and that these equilibrium points tend to cluster around the center of the phase diagram, where compositions are close to equimolar. This behavior can be explained by considering the mean initial fractions, $\bar{\phi}_i = M^{-1} \sum_n \phi_i^{(n)}$, which also cluster at equimolar compositions when $M > 1$; see Fig. S2A–D. Since the equimolar part of the phase diagram is overrepresented in the mean initial composition, we expect that also the resulting measured distribution of phases, $P(K)$, is biased toward this part of the phase diagram. This might actually be relevant to the biological example, since it demotes the large region of the phase space where one or more components are present only in trace amounts and thus could never phase separate. In any case, we use the same sampling for all interaction matrices discussed in the main text, so that comparisons of $P(K)$, and in particular the deduced performance g, are informative.
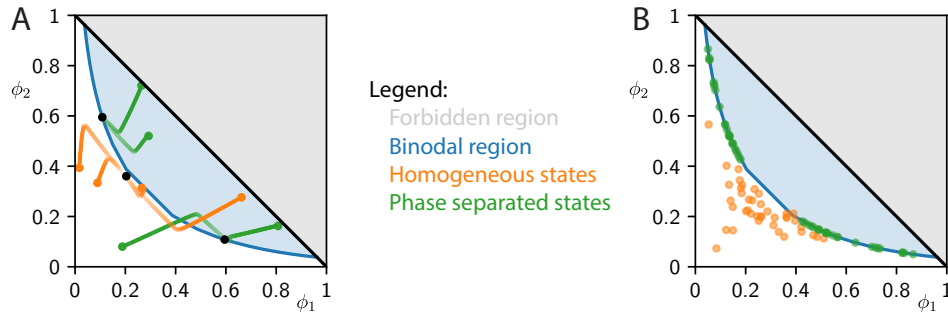
3. Supplementary figures

Fig. S1. Simplified dynamics reveal equilibrium states. A system with $N = 2$ components is investigated for $\chi_{ij} = 3.5\,(1 - \delta_{ij})$. The coexistence (binodal) line is shown in blue, while they gray region is forbidden since $\phi_1 + \phi_2 > 1$. (A) Two representative trajectories of $M = 4$ phases with random initial conditions. The four initial compositions (colored dots) evolve toward one value ($K = 1$, orange trajectories) or two different compositions ($K = 2$, green trajectories). (B) Final compositions of 64 runs with random initial conditions. The final states are colored according to whether the $M = 4$ phases all obtain the same composition ($K = 1$, orange dots) or two different compositions ($K = 2$, green dots). Note that all green dots lie on the binodal line (blue), while all homogeneous states (orange dots) are outside the binodal region.
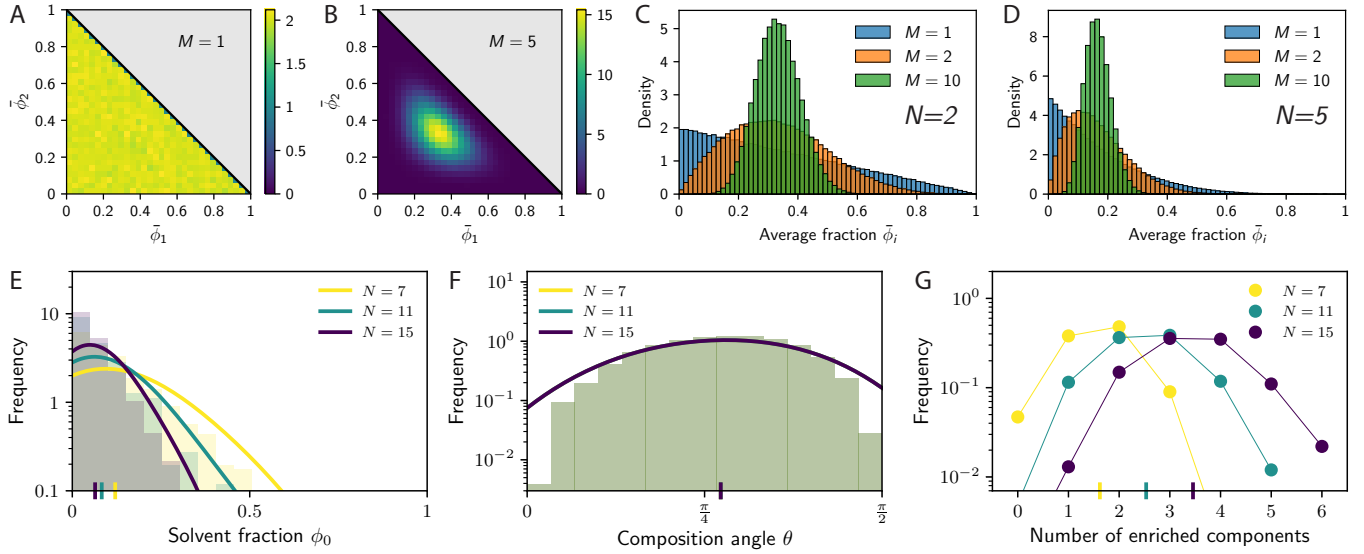
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Fig. S2. Uniform initial compositions exhibit a large variability. (A, B) Density of mean fraction $\bar{\phi}_i = M^{-1} \sum_n \phi_i^{(n)}$ for uniformly chosen $\phi_i^{(n)}$ for $N = 2$ and various number of phases, M. (C, D) Density of $\bar{\phi}_i$ of each component i for various N and M. (E) Histograms and kernel density estimates of the solvent fraction ϕ_0 for various component counts N. Since the composition is uniformly distributed, the fraction of each component follows the given distribution. (F) Histograms and kernel density estimates of the composition angle θ for N, suggesting that this distribution is independent of N. (G) Distribution of the number of components enriched in phases for various N. (E–G) The means of the distributions are indicated by vertical bars on the horizontal axes.
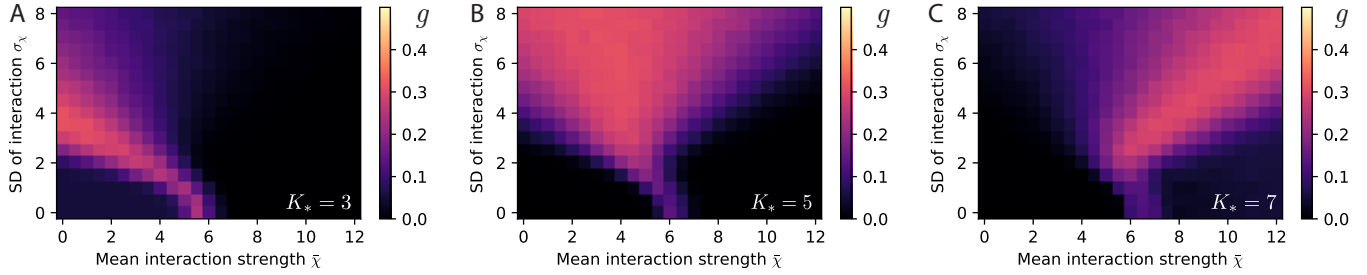

Fig. S3. Random interaction matrices perform sub-optimally. (A–C) Performance g of interaction matrices χ_{ij} as a function of their mean $\bar{\chi}$ and standard deviation σ_χ for $N = 9$ components for target phase counts $K_* = 3, 5, 7$ and $w = 1$.
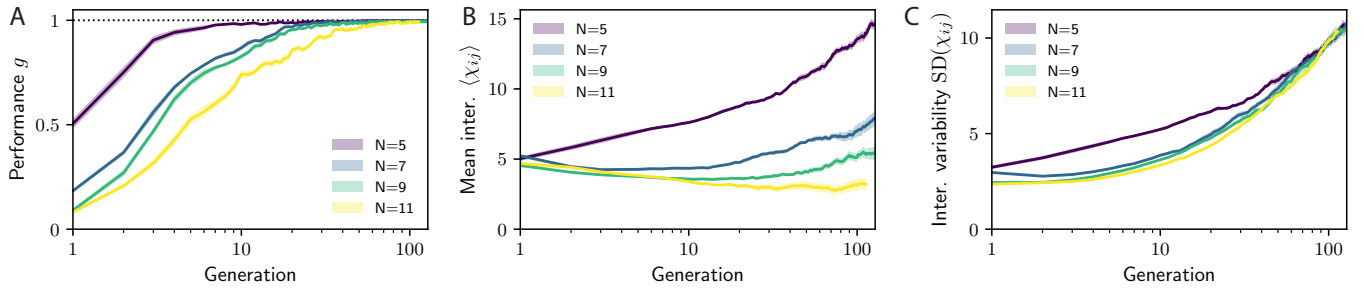

Fig. S4. Unconstraint evolution obtain optimal solutions at the expense of unphysical large interactions. (A) Performance g as a function of generation for different number of components N. (B) Interaction strength $\langle \chi_{ij} \rangle$ as a function of generation for various N. (C) Associated standard deviation $\mathrm{SD}(\chi_{ij})$ as a function of generation for various N. (A–C) Additional model parameters are $\sigma_e = 1$, $K_* = 5$, and $w = 1$.
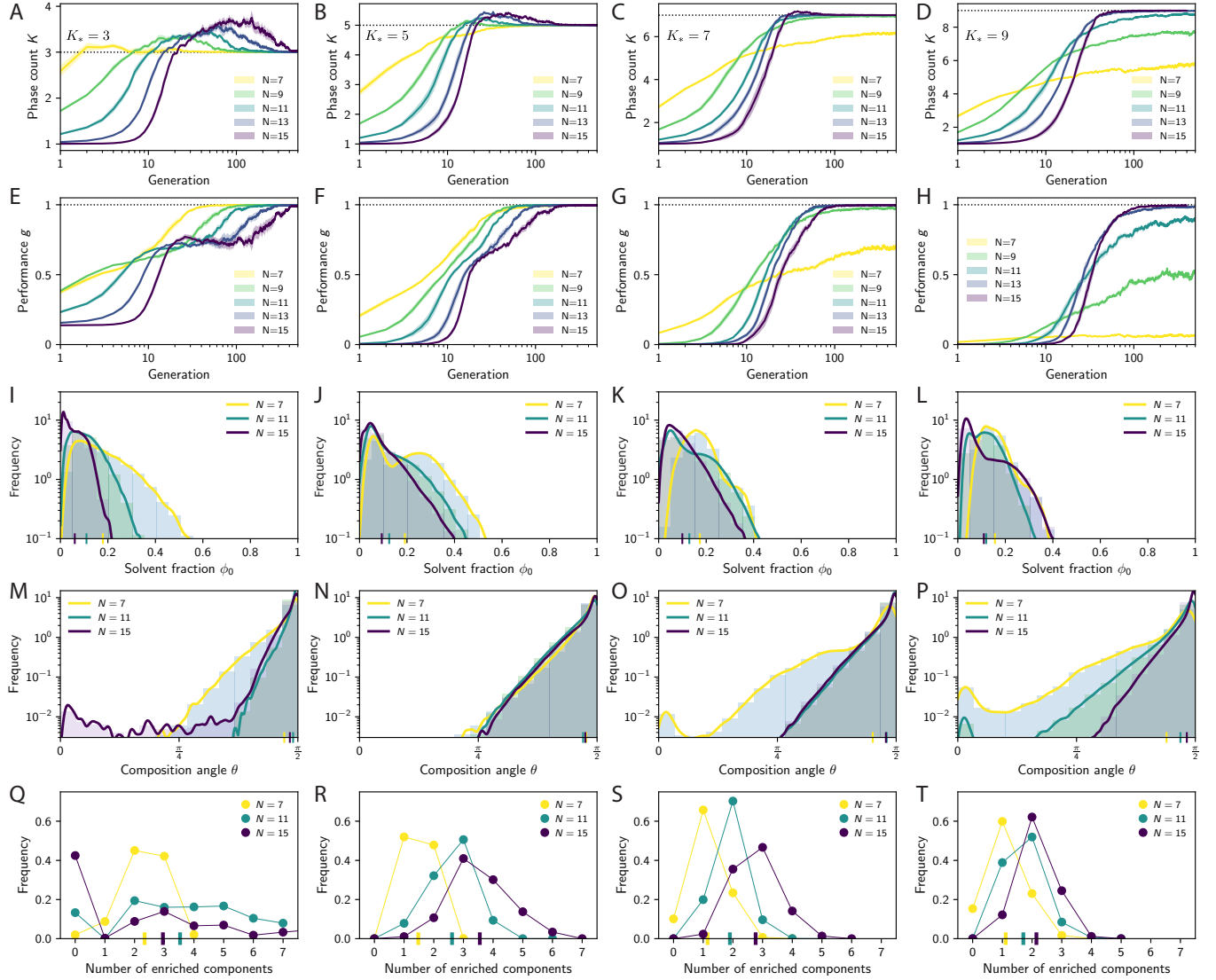
Fig. S5. Evolution of interaction matrices for various target phase counts K_* (columns) and different number of components N (colors); The second column ($K_* = 5$) corresponds to Fig. 4 in the main text. (A–D) Phase count K as functions of generation. K_* indicated by a dotted line. (E–H) Performance g as a function of generation. Maximal performance indicated by dotted line. (I–L) Distribution of the solvent fraction ϕ_0 shown as histograms and using kernel density estimation (lines). (M–P) Distribution of composition angles θ shown as histograms and using kernel density estimation (lines). (Q–T) Distribution of the number of components enriched in phases. (A–T) Bars on the x axes indicate distribution means. Additional model parameters are $w = 1$, $\sigma_{\mathrm{e}} = 0.3$, and $\chi_{\mathrm{bound}} = 5$.
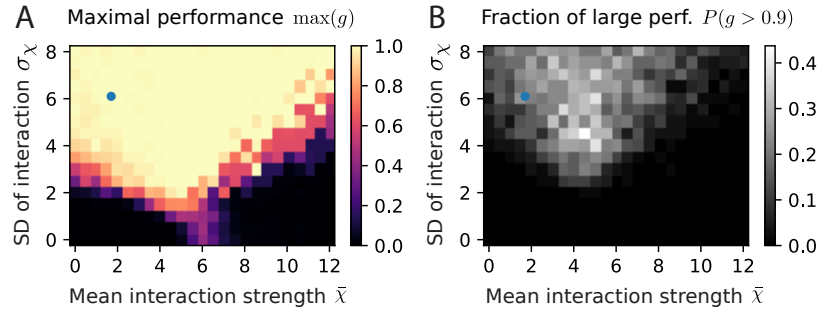
Fig. S6. Optimal matrices are frequent in the random ensemble. Maximal performance g (A) and frequency of $g > 0.9$ (B) as a function of the mean $\bar{\chi}$ and standard deviation σ_χ of normally distributed interactions χ_{ij}. The blue dot indicates the statistics of the evolved matrices (Fig. 4E in the main text). 64 random matrices have been considered for each pair $(\bar{\chi}, \sigma_\chi)$. Additional parameters are $N = 9$, $K_* = 5$, and $w = 1$.

Movie S1. Evolution of an interaction matrix during optimization. The video shows how a representative interaction matrix from the evolutionary optimization shown Fig. 4 evolves over 500 generations.

References

1. F Jülicher, SW Grill, G Salbreux, Hydrodynamic theory of active matter. *Rep. Prog. Phys.* **81**, 076601 (2018).
2. JW Gibbs, On the equilibrium of heterogeneous substances. *Trans. Conn. Acad. Arts Sci.* **3**, 1–329 (1876).