Glass Transition and Lack of Equipartition in a Statistical Mechanics model for Random Lasers

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Recent experiments suggest that the onset of lasing in optically active disordered media is related to an ergodicity-breaking transition for the degrees of freedom of the electromagnetic field. We test this hypothesis in numerical simulations of the dynamics of nonlinearly coupled light modes under external pumping. The collective behavior of light mode amplitudes appears to be akin to the one displayed in glass formers around the ergodicity breaking glass transition: a critical pumping exists, beyond which the thermodynamic phase is fragmented into a multitude of states. The probability distribution of the overlap between such states, i.e., the glass order parameter, turns out to be well described by the replica symmetry breaking scheme. The unprecedented observation is that such symmetry breaking occurs at the same pumping power values at which a lack of equipartition among light modes arises. Finally, we show that the mean-field scenario for the glass transition is quite robust for the description of the physics of random lasers.

Recent experiments on optically-active disordered media, displaying random lasing above a given pumping threshold, have provided the evidence of particularly non-trivial correlations between shot-to-shot fluctuations\textsuperscript{[1–7]}. A statistical analysis of the distribution of such fluctuations shows that, crossing the lasing threshold, their variance has a fast accelerating increase with the power. A further probe of the distribution of the similarities of pairs of fluctuations in different shots, termed intensity fluctuation overlaps (IFO’s), has shown that in such compounds strong correlations among fluctuations are compatible with an organization of emission mode configurations in clusters of states, akin to the one occurring for the multitude of thermodynamic states composing the glassy phase in glass formers. Such a correspondence has been theoretically explained proving the equivalence between the distribution of IFO’s and the distribution of the overlap between states, the so-called Parisi overlap, that is the order parameter of the phase transition in glassy systems. The analytical proof has been, though, derived assuming very narrow-band spectra, such that all modes can be considered at the same frequency. This is not the case, however, for many realistic multimode lasers, both ordered and random. In particular, a non-linear feature of standard multimode lasers - providing short pulses - is mode-locking. A necessary condition for mode-locking to occur is that a matching condition is satisfied among the frequencies of sets of coupled modes.\textsuperscript{[4–6]} E.g., in the four-wave mixing case\textsuperscript{[4–6]} theory predicts that given four waves of indices $i_1, i_2, i_3, i_4$ they can be actually coupled if and only if their frequencies satisfy $|\omega_{i_1} - \omega_{i_2} + \omega_{i_3} - \omega_{i_4}| < \gamma$; $\gamma$ being the typical line-width of a mode. It has been recently experimentally demonstrated that mode-locking occurs in the GaAs powder random laser\textsuperscript{[8]}.

The above observations make the investigation of multimode models of random lasers a necessary step to understand the fundamental mechanisms at the ground of the fascinating phenomenon of lasing in random media. The first goal of this work is to undertake such investigation by means of numerical simulations and bridge the (random) laser phenomenology to the one of critical phenomena in glassy systems. In particular, the question we want to answer is whether the narrowing of the emission spectrum experimentally observed at the lasing threshold and the related crossover to coherent light emission are due to an underlying phase transition. In panel a) of Fig.\textsuperscript{1} we show an instance of spectra of ZnO nanoparticles in a rhodamine 640 dye solution, taken from\textsuperscript{[9]}. We will show that the phase transition scenario is correct: a glass transition takes place for the degrees for freedom of the electromagnetic field in the optically active disordered medium.

Another goal of the present work is to bridge the phenomenology of ergodicity breaking in systems with coupling frustration and in non-linear systems with ordered interactions, as for instance the famous Fermi-Pasta-Ulam (FPU) model\textsuperscript{[10]}, a one dimensional chain of classical particles coupled by non-linear springs. For the FPU model the first numerical simulation ever performed, in Los Alamos in 1955, showed that, taking an initial condition far from equilibrium, there is a non-ergodic regime persisting on very long time-scales. Such a regime is typically revealed as a persistent lack of equipartition between the fundamental degrees of freedom of the system\textsuperscript{[11–16]}. This kind of ergodicity breaking phenomenology, which is also typical of non-linear systems displaying breathers or solitonic

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\textsuperscript{1} This is the dominant term in absence of non-centrometric, non- time translational invariant phenomena like double harmonic generation. That is, when $\chi^{(2)}$ contributions to non-linear optical susceptibility are negligible with respect to $\chi^{(3)}$. 

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excitations [17–19], was never observed, to our knowledge, in systems with disordered interactions in their glassy phase.

The present work is the first one focused on a model where the lack of equipartition between the fundamental degrees of freedom and the non-trivial features of the Parisi overlap distribution appear at the same time, strongly suggesting that these are just two complementary way to detect the same underlying phenomenon: the breaking of ergodicity. This observation is supported by our numerical data on a statistical mechanics model for non-linear waves interaction in random media, the mode-locked 4-phasor model. These kind of models with random couplings, pertaining to the set of p-spin models, are not usually studied in numerical simulations, because of their high computational cost, but are used to carry out analytical computation. In that context they are never studied in inhomogeneous interaction networks, such as the one produced by the frequency matching condition among light mode frequencies, which on the contrary characterizes the mode-locked 4-phasor model studied here. In the random laser case the 4-phasor model emerges naturally to describe mode dynamics in the stationary regime [20–24]. Since the model variables - complex continuous - can change their magnitude, unlike Ising, XY or Heisenberg spins, one can contemporarily probe energy equipartition among degrees of freedom and the occurrence of a phase transition. This is, thus, the first unprecedented example of a model where lack of equipartition and state fragmentation at the critical point appear as concomitant effects.

RESULTS

Model

We study a statistical mechanics model for the non-linear interactions of the electromagnetic field in a disordered optically active medium: the mode-locked 4-phasor model. The phasors, dynamic variables of the system, are the complex amplitudes $a_k(t) = A_k(t) e^{i\phi_k(t)}$ of the electromagnetic field expansion in normal modes $E(r, t) = \sum_k a_k(t) e^{i\omega_k t} E_k(r) + c.c.$. Being $A_k = |a_k| \in \mathbb{R}^+$ and $\phi_k = \arg a_k \in [0, 2\pi]$, the dynamics of the stationary regime can be shown to be a stochastic potential dynamics whose Hamiltonian reads [21, 25]:

$$H[a] = -\sum_{\vec{\tau}} J_{\vec{\tau}} A_{i_1} A_{i_2} A_{i_3} A_{i_4} \cos(\phi_{i_1} - \phi_{i_2} + \phi_{i_3} - \phi_{i_4}),$$  \hspace{1cm} (1)

where the coefficients $J_{\vec{\tau}} = \{i_1, i_2, i_3, i_4\}$ are quenched random variables (we will take them Gaussian) and the $\sum_{\vec{\tau}}$ specifies a particular light mode network. We also implement gain saturation into the model, that can be formally rephrased into a constraint on the total intensity: $\epsilon N = \sum_{k=1}^{N} A_k^2$ [20–24]. Despite the fact that energy is continuously injected and dissipated within a random laser, according to [20–21–24–25] one can assume an effective equilibrium distribution for the amplitudes:

$$P(a_1, \ldots, a_k) = e^{-\beta H[a]} \delta \left( \epsilon N - \sum_{k=1}^{N} A_k^2 \right),$$  \hspace{1cm} (2)

where $\beta$ is some effective inverse temperature and $\epsilon$ measures the optical power per mode available to the system. Rescaling $A_k \rightarrow A_k / \sqrt{\epsilon}$ in Eq. (2), the new variables are constrained on the same hypersphere at the cost of a rescaling of the effective temperature as

$$\beta \rightarrow \beta \epsilon^2 = \mathcal{P}^2$$  \hspace{1cm} (3)

where $\mathcal{P}$ is the so-called pumping rate parameter. In these rescaled variables the Boltzmann weight reads

$$\rho(\{a\}) = \frac{1}{Z} \exp\{-\mathcal{P}^2 H[a]\},$$

making explicit the role of the pumping as effective heat bath for the stationary regimes of the lasing random media.

We have investigated how the system behaves varying the pumping rate $\mathcal{P}$. According to Eq. (2) and Eq. (3), in numerical simulations it is identical to fix the constraint $\epsilon$ and change the temperature $T$ or work at fixed temperature
freedom, introduced in the framework of nonlinear systems [11, 13]:

\[
|\omega_{i_1} - \omega_{i_2} + \omega_{i_3} - \omega_{i_4}| \lesssim \gamma,
\]

where \( \gamma \) is the typical line width. The FMC constraint introduce in the topology of the interaction network inhomogeneities such that standard mean-field approximations used solve the thermodynamics of disordered systems [29, 30] are not exact. Let us consider for instance the simple case of a linear dispersion relation with equiparted angular frequencies, \( \omega_j = \omega_0 + j \delta \), with \( \delta \ll \gamma \) and \( i = 1, \ldots, N \). Eq. (1) very simply reads as the constraint \( |i_1 - i_2 + i_3 - i_4| = 0 \) on the summation indices in Eq. (1), diluting by a factor \( N \) the number of interactions from the fully connected case, \( N(N-1)(N-2)(N-3)/24 \) [31]. We call the resulting interaction network the mode-locked graph, whose detailed discussion can be found in the corresponding section in Methods.

**Observables**

The assumption that the stationary probability distribution of light mode amplitudes is an effective equilibrium one [see Eq. 2] allows us to use an equilibrium Monte Carlo dynamics to sample phase space. To speed up the equilibration we use the Parallel Tempering algorithm [32]. To show that the crossover from fluorescence to random lasing is an ergodicity breaking transition, we have measured standard order parameters to detect ergodicity breaking. The first, and simplest conceptually, is the spectral entropy:

\[
S_{sp} = -\sum_{i=1}^{N} \hat{I}_k \log(\hat{I}_k)
\]

\[
\hat{I}_k = \frac{\langle A_k^2 \rangle}{\sum_{k=1}^{N} \langle A_k^2 \rangle},
\]

measuring the energy degree of equipartition in the spectrum, \( \hat{I}_k \) being the normalized thermal average of the intensity for given wave number \( k \). The angular parentheses ( ) denote the thermodynamic average (see Methods). The information content of the spectral entropy can be conveniently expressed by the effective number of degrees of freedom, introduced in the framework of nonlinear systems [11, 13]:

\[
n_{eff} = \frac{\exp(S_{sp})}{N},
\]

where \( n_{eff} = 1 \) for perfect equipartition and \( n_{eff} = 0 \) when the total energy is concentrated on a finite fraction of modes. Values \( n_{eff} < 1 \) signal the breaking of equipartition [11, 13].

We also measured the order parameter introduced to detect ergodicity breaking in the context of disordered systems thermodynamics, the Parisi’s overlap [21, 22, 33, 34]:

\[
q^{\alpha \beta} = \frac{1}{N} \sum_{k=1}^{N} u_k^{\alpha} u_k^{\beta} = \frac{1}{N} \sum_{k=1}^{N} A_k^{\alpha} A_k^{\beta} \cos(\phi_k^{\alpha} - \phi_k^{\beta}).
\]

The greek indices in Eq. (8) denote different replicas, i.e., independent configurations at equilibrium at the same temperature \( T \). The Parisi’s overlap is characterized by a low temperature non-trivial distribution in presence of a glass phase [29, 33]. We have, further, introduced the overlap between the energy stored on different interactions, i.e., the plaquette overlap:

\[
Q^{\alpha \beta} = \frac{1}{N_4} \sum_{i=1}^{N_4} P_i^{\alpha} P_i^{\beta},
\]

where \( P_i \) is the plaquette degree of freedom,

\[
P_i = A_{i_1} A_{i_2} A_{i_3} A_{i_4} \cos(\phi_{i_1} - \phi_{i_2} + \phi_{i_3} - \phi_{i_4}),
\]
Figure 9. Spectra of emission from the rhodamine 640 dye solution containing ZnO nanoparticles. The ZnO particle density is $\sim 6 \times 10^{11}$ cm$^{-3}$. The incident pump pulse energy is (from bottom to top) 0.74, 1.35, 1.7, 2.25 and 3.4 $\mu$J.

As a this mode. A further increase of optical gain leads to lasing in more low-loss modes. Laser emission from these modes gives discrete peaks in the emission spectrum (figure 9). When the scattering strength increases further, the decay rates of the eigenmodes and the coupling among them continue decreasing. There are a small number of eigenmodes with extremely long lifetime and nearly decoupled from other modes. The threshold gain for lasing is $\frac{P_c}{P} = 1.71$. $1.15$, $0.97$, $0.64$.

FIG. 1. Panel a): Emission spectrum of ZnO nanoparticles in a rhodamine 640 dye solution. From bottom to top: increasing injected optical power $P$. The emission spectrum of rhodamine shown here is a reproduction of the original Fig. 9 appearing in [H. Cao, Waves Random Media 13, R1 (2003)]. Panel b): Intensity spectrum $I_\lambda = \langle A_\lambda^2 \rangle$ as a function of the wavelength $\lambda$ averaged over many instances of the quenched randomness, numerical simulations with $N = 64$ degrees of freedom. The wavelength $\lambda = 2\pi/k$ is expressed here in arbitrary units ([a.u.]). Notice the narrowing of the spectrum at larger values of the pumping rate $P$. Panel c): Intensity spectrum $I_\lambda$ as a function of the wavelength $\lambda$ for a single instance of the quenched randomness, numerical simulations with $N = 64$ degrees of freedom. Color code of the curves: $P$ increases from bright (yellow) to dark (purple). Notice the crossover from a smooth, almost equipartited, spectrum at low $P$ to a disordered pattern of isolated peaks at high $P$.

and where $N_4$ is the number of four-body couplings (plaquettes) appearing in the definition of energy in Eq. (1). We have studied the two probability distributions $P(q)$ and $P(Q)$.

As is clear from their definitions in Eqs. (9) and (10), in order to measure $P(q)$ and $P(Q)$ one needs to know the phases of the electromagnetic modes [35], which are experimentally not retrieved so far in random lasers [8, 36–38]. For this reason it was introduced and measured in [1, 27] the so-called Intensity Fluctuation Overlap (IFO), an ergodicity
FIG. 2. **Panel a):** Effective number of degrees of freedom \( \bar{n}_{\text{eff}} \) (disorder average) as a function of the inverse (squared) pumping rate \( P^{-2} = T \), corresponding to the temperature in the numerical algorithm. \( \bar{n}_{\text{eff}} \approx 1 \) signals equipartion, \( \bar{n}_{\text{eff}} < 1 \) lack of equipartition. **Panel b):** Binder parameter \( B \) for the probability distribution of \( n_{\text{eff}} \), measured as a function of \( P^{-2} \). **Panel c):** Bimodality parameter \( b \) for \( P(n_{\text{eff}}) \) measured as a function of \( P^{-2} \). **Panel d1) & d2):** Spectra obtained from different instances of the disorder are shown for the equipartited phase, \( (P_c/P)^2 \approx 2 \), in **d1**, and the non-equipartited phase, \( (P_c/P)^2 \approx 0.5 \), in **d2**.

breaking order parameter for complex waves amplitudes which do not depend on phases:

\[
C^\alpha\beta = \frac{1}{N} \sum_{k=1}^{N} \Delta^\alpha_k \Delta^\beta_k
\]

\[
\Delta^\alpha_k = \frac{A^\alpha_k - \langle A^\alpha_k \rangle}{2\sqrt{2\epsilon}}
\]

where the indices \( \alpha \) and \( \beta \) label independent equilibrium configurations. From the point of view of an experiment the idea is to consider, for a given experimental setup (quenched randomness) and a fixed value \( P \) of the pumping rate, the average value \( \langle A^\alpha_k \rangle \) for each line of the spectrum and then study the shot-to-shot, i.e., the replica-to-replica, correlation between fluctuations around this average value. We have, thus, numerically measured the distribution \( P(C^\alpha\beta) \).

All the above mentioned distributions have been characterized by measuring their Binder parameter \( B = (3 - \kappa)/2 \) and their bimodality parameter \( b = (\gamma^2 + 1)/\kappa \), where \( \kappa \) and \( \gamma \) are respectively kurtosis and skewness (more details in Methods).

**Breaking of equipartition**

We first characterize the behaviour of the intensity spectrum vs the pumping rate \( P \). As reported in panel a) of Fig. 1 for one instance of experimental measurements \[39\], a narrowing of the spectrum is commonly observed as \( P \) increases.
The spectra computed in our numerical simulations, shown in panel b) of Fig. 1, display the same behaviour. Besides the possibility of tuning at will the control parameters of the system, one of the great advantages of numerical simulations is the possibility to look at the spectrum dependence on the temperature for a single instance of disordered couplings. In panel c) of Fig. 1 we see that increasing $P$ a crossover occurs from an almost equipartitioned regime to a regime where energy is concentrated on some peaks arranged in a disordered pattern. Data from a single instance of disorder, thus, suggest that the spectral narrowing accompanying the onset of random lasing in experiments [31, 37, 39] has to be related to a breaking of equipartition. The degree of equipartition is characterized by the effective number of degrees of freedom $n_{\text{eff}}$ [see Eq. (7)], a parameter introduced in the FPU literature to detect detect non-ergodicity through the lack of equipartition [11, 13, 15].

In panel a) of Fig. 3 we show the behaviour of $\pi_{\text{eff}}$, the average over different instances of the disordered couplings, as a function of the pumping rate $P$. We plot data as function of the squared inverse pumping rate, that is, the temperature $T = P^{-2}$. At low pumping rate (high $T$) we find a good degree of equipartition between electromagnetic field modes, $\pi_{\text{eff}} \approx 1$, for all the sizes $N$ studied. By increasing the pumping rate (decreasing $T$) $\pi_{\text{eff}}$ drops to lower values at a breaking point which slightly depends on $N$. We can roughly identify this breaking point as the lasing threshold $P_c(N) = \sqrt{\langle T_c \rangle}$ for the pumping rate. By looking at the curves in panel a) of Fig. 1 it is clear that the decrease of $\pi_{\text{eff}}$ is the steeper the larger is $N$: in the framework of critical phenomena this is the first indication that the crossover to non-equipartition is a first-order transition. A stronger evidence of the first-order nature of the transition comes from the study of the Binder parameter $B$ and of the bimodality parameter $b$ of the distribution $P(n_{\text{eff}})$ varying $P$, shown respectively in panel b) and panel c) of Fig. 2. The value $P_c(N)$ estimated as the point where $B$ signals a maximal deviation from Gaussianity corresponds to the breaking point of $\pi_{\text{eff}}$, as well as to the peak of the bimodality indicator $b$. The last observation confirms that the deviation from Gaussianity is due to a bimodal nature of the $n_{\text{eff}}$ distribution, thus confirming the first-order transition scenario [40]. We believe that to reproduce such an analysis in experiments would be of primary interest. Finally, in order to emphasize the difference in the degree of equipartition at different values of $P$, in panel d) of Fig. 2 spectra obtained for different instances of the random couplings are compared at low pumping rate, panel d1), and high pumping rate, panel d2).

As we already mentioned, in the context of non-linear systems the lack of equipartition is usually regarded as a landmark of ergodicity breaking [11, 16], often also related to the appearance of “localized objects” as breathers or solitons [17, 19]. But for that class of systems there is no example showing that the lack of equipartition is also related to a non trivial distribution of the overlap order parameter introduced by Parisi 40 years ago to characterize ergodicity breaking in disordered systems [29]. The goal of the next section is to show that, on the contrary, for the mode-locked 4-phasors model this is precisely what happens: the lack of equipartition takes place together with, following the terminology of [29], the breaking of the symmetry between replicas.

Glass transition

As promised, we now investigate to which extent the narrowing of the emission spectrum, see Fig. 1 is related to a glass transition. More precisely, to the so-called Random First-Order Transition (RFOT) [11, 24]: a mixed-order transition with no latent heat but with a discontinuous order parameter. This is the overlap distribution $P(q)$ changing from a single peaked distribution to a bimodal one at the critical point, a behaviour typical of first-order transitions. In the high $T$ ergodic phase the distribution $P(q)$ is peaked at the origin $q_0 = 0$. Below the glass transition temperature $P(q)$ develops a secondary peak at a finite distance $q_1 > q_0$ from the origin [23]. This behaviour is typical when phase space splits into disjoint ergodic components. The overlap of configurations inside the same ergodic component is $q_1$ while the mutual overlap of configurations belonging to different ergodic components is $q_0$. Let us show that the numerical results on the mode-locked 4-phasor model are in complete agreement with this scenario.

In panels a) and b) of Fig. 3 we display the specific-heat per mode

$$C_V = \frac{\langle E^2 \rangle - \langle E \rangle^2}{N T^2}$$

vs $T = 1/P^2$ for different system sizes. The overbar denotes the average over disordered couplings realizations, whereas the angular parentheses $\langle \rangle$ denote the thermodynamic average (see Methods). Besides the numerical data for the model on the mode-locked graph we also report, as a comparison to standard RFOT systems, numerical data obtained for another similar system, the random diluted 4-phasor model. The latter is defined by the same variables and interactions of the mode-locked 4-phasor model, but lives on a different graph, characterized by the same number of non-zero interactions and the same scaling with $N$ as the mode-locked one, but obtained by randomly diluting a fully connected graph in which each mode interacts in every possible set of four modes (see Methods for details).

At all sizes $C_V/N$ has a characteristic non-monotonic behaviour, with a peak at a $T$ that depends on $N$. We identify this point as a finite size “critical temperature” $T_c(N) = 1/P^2$. The good collapse of the curves at different
In panel a) of Fig. 3, the randomly diluted graph, two pronounced shoulders at identical system size, i.e., with the same number of modes, develop in peaks at larger temperatures $T/T_c(N) = 1.71$, $1.15$, $0.97$, and $0.64$. The scaling regime of the specific heat is, here, $\sim (T/T_c-1)^{\nu} N^{\gamma}$, where $\tau = T/T_c(N) - 1$, curve collapse in the scaling region. The four sizes are $N = 32, 48, 64, 102$.

Panel b): 4-phasor model on the randomly diluted graph. The scaling regime shrinks with size as $\tau \sim N^{-3/2}$. The scaling exponent $1/\nu = 3/2$ deviates remarkably from mean-field [1.4], which would predict $1/\nu = 1/2$ for a theory of second-order transition with quartic non-linearity. As a comparison, in panel b) of Fig. 3, the curves of the randomly diluted 4-phasor model (with the same amount of dilution of the mode-locked one) are displayed. The scaling regime of the specific heat is, here, $\tau \sim N^{-1/2}$, in agreement with a simple mean-field model of the glass transition [1.1]. The strong deviation from mean-field scaling in the mode-locking case is due to the correlated way in which the interaction network is diluted and mainly depends on the fact that, because of FMC, Eq. [4], modes whose frequencies are at the center of the spectrum tend to interact more than modes whose frequencies are at the boundaries. Though this effect is expected to vanish in the thermodynamic limit $N \rightarrow \infty$, this very strong finite size effects reflect typical features of random laser compounds, such as the spectral narrowing with $P_c$.

In panel c) and d) of Fig. 3 we display the distribution of the modes overlap $P(q)$, respectively for the randomly diluted 4-phasor model [panel c] and the mode-locked model [panel d], across the critical scaling region. For both models the probability distribution $P(q)$ is a Gaussian centered on the origin at low pumping rate $P$. In the case of random dilution, above the critical value $P_c(N)$ one finds secondary peaks at a finite distance from the origin: they are the signature of the broken ergodicity phase. In the mode-locked graph, on the contrary, at high pumping rates we just find two pronounced shoulders at identical system size, i.e., with the same number $N$ of modes. Although shoulders at small $N$ might develop in peaks at larger $N$, it would hard to demonstrate the presence of an ergodicity-broken phase solely on the basis of the data in panel d) of Fig. 3. In mean-field models the information content of variable
Therefore, we consider, as well, the distribution and interaction overlaps is theoretically the same, though finite size corrections may depend on the observables. We cannot therefore rely on observables which require the measurement of phases. The modes and plaquette overlaps high-power directional impulses: unfortunately this is not the operating regime of standard random lasers and cannot be measured in real random lasers.

The Intensity Fluctuation Overlap

We have presented the numerical evidence that in a statistical mechanics model for random lasers there is a RFOT glass transition concomitant with the lack of equipartition. One problem is to which extent this picture can be assessed even in experiments. The available technology for the measurements of light mode phases applies only to high-power directional impulses: unfortunately this is not the operating regime of standard random lasers. We cannot therefore rely on observables which require the measurement of phases. The modes and plaquette overlaps defined Eqs. (8) and (9) depend on phases, so they are not useful for current experiments. A further intensity fluctuation overlap \( C_{\alpha,\beta} \), cf. Eq. (11), can be introduced, that - at the mean-field level - is proved to be in a one-to-one correspondence with the standard overlap, \( C_{\alpha,\beta} \propto q_{\alpha,\beta}^2 \).  

In panel b) of Fig. 4 is shown the distribution \( P(C) \) for \( N = 64 \) at four different values of the pumping rate \( P \), two above and two below the critical \( P_c \), as determined from the caloric curve (Fig. 2). At first sight there is no clear evidence of secondary peaks at high pumping rates for this system size, although non-Gaussian tails appear in the vicinity of the transition. It is the study of the Binder parameter \( B \) dependence on \( P \) which reveals how \( P(C) \) brings the signature of a first-order transition: data are shown in the inset of panel b) of Fig. 4. The behaviour of \( B \) as a function of \( P \) (plotted as \( B \) vs \( P^{-2} = T \) to have a clearer term of comparison with the literature) is the one characteristic of first-order transitions. This behaviour of the IFO order parameter is, therefore, also consistent with the RFOT scenario and with the study of the breaking of equipartition in the spectrum: in all the three cases the ergodicity-breaking parameter behaves as the order parameter of a first-order transition.

**CONCLUSIONS**

By means of Monte Carlo numerical simulations of a statistical mechanical model of the non-linear interactions of light in a random medium, we have shown that the onset of random lasing has the properties of a glassy phase transition. More precisely it is an ergodicity-breaking transition characterized by a diverging second order susceptibility (the specific-heat) and an order parameter, the overlap, with first-order features at the transition. As weird as it can sound, light multiply scattering in random media really looks like, among physical systems, a very good benchmark.

**FIG. 4. Panel a)** Plaquette overlap distribution \( P(Q) \) for \( N = 64 \) and \( N_t = 2^{14} \). Inset: Multimodality parameter \( b \) measured for \( P(Q) \), values above the threshold \( b^* = 5/9 \) (full black line) indicate a bimodal distribution. **Panel b)**: Intensity Fluctuation Overlap (IFO) probability distribution \( P(C) \) for system size \( N \) at \( N_t = 2^{14} \), four different values of the pumping rates: \( (P_c/P)^2 = 1.71, 1.15, 0.97, 0.64 \). Inset: Binder parameter \( B \) measured for \( P(C) \) as a function of \( P^{-2} = T \); the behaviour is the typical one of first-order transitions, with the transition at the minimum of \( B \).
to test the existence and the hidden nature of a glass transition.

Further on, thanks to this feature and to the properties of the model adopted, we were able to show unprecedented evidence of a deep connection between lack of equipartition - typical of ergodicity breaking in non-disordered systems with non-linear interactions [10, 11, 15] - and the breaking of ergodicity as described within the paradigm of replica-symmetry-breaking [30]. The mode-locked 4-phaser model is the first example of a system where ergodicity breaking manifest itself at the same time as a breaking of the symmetries between replicas and as a lack of equipartition. This important result suggest a possible way to overcome the intrinsic difficulty usually encountered in the measure of $q$, which is not a single-experiment observable. The standard protocol is that one needs to compare the results of several experiments done on the same sample or of several numerical simulations with the same realization of quenched disorder to obtain a measure $P(q)$. Using the jargon of disordered systems one needs more replicas of the same system. As we showed, when the occurrence of a non-trivial $P(q)$ is simultaneous with the loss of spectral equipartition, one can simply take advantage of latter to detect the ergodicity breaking glass transition.

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METHODS

The Mode-Locked graph

The first step of the numerical study is the generation of the mode-locked graph. It is easier to describe the structure of the interaction network as a bipartite graph where interaction nodes $J_\mu$ labeled by greek letters are connected to variables nodes $A_k$ labeled with latin letters. Since we have a four-body interaction each interaction node is always attached to 4 variable nodes and is defined by the ordered list of their indices $J_\mu(i, j, k, l)$. The order is relevant because from the point of view of the energy stored in the interaction (see Eq. (1)) there are non-equivalent permutations. The steps to generate the mode-locked graph are as follows:

1. a virtual complete graph with $N!/(N-4)!4!$ interaction nodes is generated;

2. for each interaction node the three non equivalent permutations are considered: $P_\mu(i_1, i_2, i_3, i_4)$, $P_\mu(i_2, i_1, i_3, i_4)$ and $P_\mu(i_1, i_2, i_4, i_3)$; each time a non-equivalent permutation satisfies the FMC, the corresponding interaction of the virtual graph is added to the real graph.

3. The procedure at point 2 is repeated until a preassigned number of interactions in the complete graph is reached, for computational reasons this number is always a power of 2.

For large $N$, the above procedure tends to cut $O(N)$ of all interacting quadruplets [31]. Operatively, for a system with $N$ complex variables we have drawn a bipartite graph with a number of interactions scaling as $O(N^3)$ and equal to the power of 2 soon smaller than the number of all possible interactions fulfilling the FMC constraint. The number of interaction nodes $N_\alpha$ corresponding to each $N$ is listed in Tab. I.

Concerning the structure of the topology of the interaction network it is important to stress that the dimensionality of the disordered optical medium in real space is scarcely important for the thermodynamics of the problem: the interaction among modes remains in any case highly non-local in the basis of normal modes. The only quantity which depends on the real-space dimensionality is the spatial overlap between the normals modes, the information about which is stored in the disordered coefficients $J_\ell$ of the Hamiltonian in Eq. (1) as $[21, 26]$

$$J_{ijkl} = \frac{i}{2} \omega_{i1} \omega_{i2} \omega_{i3} \omega_{i4} \int_V \mathcal{N}^{(3)}(q_1, q_2, q_3, q_4; r) E_{\alpha_1}^{\alpha_1}(r) E_{\alpha_2}^{\alpha_2}(r) E_{\alpha_3}^{\alpha_3}(r) E_{\alpha_4}^{\alpha_4}(r).$$ (13)
where $\chi^{(3)}_i$ is the non-linear susceptibility of the system. Without any loss of generality we assumed such couplings to be independent Gaussian random variables with zero mean and variance $(J^2) \sim N^{-2}$, which guarantees energy extensivity.

In random lasers couplings as expressed in Eq. (13) are, in general, disordered because modes display different spatial shape and extension [43, 49]. The constituents of the integrals in Eq. (13) are very difficult to calculate from first principles. The only specific form of the non-linear susceptibility has been computed by Lamb [50] for few-mode ordered lasers and no analogue study for RLs has been performed so far, to our knowledge. Integrals like Eq. (13) in a random medium can be regarded as a sum over many random variables. Different couplings involving a given mode might, in general, be correlated [52]. Since, however, we are interested in the critical behavior, thus in the large size limit of our simulated systems and since correlations decay with the size of the system, we adopt as working hypothesis a Gaussian distribution for each $J_i$:

$$P(J_i) = \sqrt{\frac{N^2}{2\pi}} \exp\left\{ -\frac{N^2J_i^2}{2} \right\}$$ (14)

We further stress that, from the perspective of probing RFOT, considering correlated $J$'s leads to qualitatively analogue phase diagram as it is well known in spin-glass systems such as, e.g., the Random Orthogonal Model [53, 54].

If we look at interactions in the space of normal modes the system is infinite-dimensional: any degree of freedom participates to $O(N^2)$ interactions. That is why we expect the mean-field glass transition scenario drawn in [21, 27] to be quite robust for the mode-locked $p$-phasor, even if the hypothesis of narrow-band is removed.

Last but not least, the non-locality of interactions between light modes also guarantees that phenomena like energy localization, a pathology of sparse networks [28, 55], are avoided.

### Numerical Algorithm

We have studied systems with $N$ complex variables $a_k = |A_k|e^{i\theta_k}$ interacting with Hamiltonian in Eq. (1). The sampling of the probability distribution in Eq. (2) was done by means of a Parallel Tempering Monte Carlo algorithm (PT). In the PT algorithm one runs $M$ simulations with local Metropolis dynamics for identical replicas of the same system, i.e. with the same quenched disorder $J_i$. For each of the $M$ copies the equilibrium distribution $e^{-\beta_i E}$ is sampled with a local Metropolis algorithm. We have runned $K$ independent simulations at temperatures $T_i = \beta_i^{-1}$, from $T_0 = T_{\text{min}}$ to $T_{N_{PT}} = T_{\text{max}}$. Each 50 steps of the local Metropolis algorithm an exchange of configurations among simulations running at neighbouring temperatures is proposed. That is, if the Metropolis algorithm for $a_i$ runs with $\beta_i$ and that of $a_j$ with $\beta_j$ one tries the following attempt: $\{ (a_i, \beta_i); (a_j, \beta_j) \} \rightarrow (a_i, \beta_i); (a_j, \beta_j)$, which is accepted with probability $p_{\text{swap}}$:

$$p_{\text{swap}} = \min\left[ 1, \frac{e^{-\beta_i H[a_i] - \beta_j H[a_j]}}{e^{-\beta_i H[a_i] - \beta_j H[a_j]}} \right]$$ (15)

The replica exchange update is proposed sequentially for all pairs of neighbouring temperatures $\beta_i$ and $\beta_{i+1}$. For all simulations the $N_{PT}$ temperatures where taken with a linear spacing in $\beta$, i.e. $\beta_{i+1} = \beta_i + \Delta\beta$.

In the local Metropolis algorithm the configuration of complex “spins” $(a_1, \ldots, a_N)$ is updated with attention to keep $\sum_k |a_k|^2 = \text{const}$. In order to fulfill this constraint each update is realized choosing at random two spins $a_i = A_i e^{i\phi_i}$ and $a_j = A_j e^{i\phi_j}$ and proposing un update to $a_i'$ and $a_j'$ such that:

$$|a_i|^2 + |a_j|^2 = |a_i'|^2 + |a_j'|^2$$ (16)

This is simply achieved by extracting three random numbers $A_i', A_j'$ and $\theta$ with uniform probability in the interval $[0, 2\pi]$ and proposing the four simultaneous updates $A_i \rightarrow A_i'$, $A_j \rightarrow A_j'$, $A_i \rightarrow A_i \cos(\theta)$ and $A_j \rightarrow A_j \sin(\theta)$, with $A_i^2 = A_i'^2 + A_j^2$. For the parallel tempering we used 32 temperatures for all sizes. We have simulated systems at four different sizes, $N = 32, 48, 64, 102$. The update of the the variables $A_i$ must be done sequentially, since the interaction network is dense and there is no way to partition the variables in subsets which can be updated independently.

We have implemented parallelization on GPU graphic cards at two levels. In order to accept or reject the update

\footnote{We do not consider contributions from the first non-linear term $\chi^{(2)}_i$ in the polarization expansion because of the relatively limited band width of the emission spectra in random lasers and the consequent lack of second harmonic generation, but that term can be easily inserted leading to no qualitative difference in the results.}
of two spins $a_i$ and $a_j$ one has to compute the energy update on $O(N^2)$ quadruplettes, i.e. $\Delta E = \sum_{k=1}^{\tilde{N}} \Delta E_k$, with $\tilde{N} = O(N^2)$. The execution of the $M$ simulations at different temperatures is implemented in parallel on the GPU. We have run simulations on two type of graphic cards: GTX 680, K 20. The best overall speedup achieved with respect to the sequential code on CPU is of a factor 8.

Binder and Bimodality parameters

The quantitative indicators used to characterize the order parameter distributions are the Binder parameter $B$ and the multimodality parameter $b$. Both these indicators are built from cumulants of the distribution. In particular one needs the definition of curtosis $\kappa$,

$$\kappa = \frac{\langle (\Delta q)^4 \rangle}{\langle (\Delta q)^2 \rangle^2}, \tag{17}$$

and skewness $\gamma$,

$$\gamma = \frac{\langle (\Delta q)^3 \rangle}{\langle (\Delta q)^2 \rangle^{3/2}}, \tag{18}$$

where $\Delta q = q - \langle q \rangle$. The angle parentheses denote thermal average while the overline denotes average over disorder realizations. The Binder parameter is $B = (3 - \kappa)/2$ and the multimodality parameter reads as $b = (\gamma^2 + 1)/\kappa$. In the case of the distribution obtained from a sample with $n$ data, which is the case considered here, the definition of the multimodality parameter is

$$b = \frac{\gamma^2 + 1}{\kappa + \frac{3(n-1)^2}{(n-2)(n-3)}}. \tag{19}$$

Replicas and Overlap

In the analytic calculation of disordered systems free energy one needs to consider the analytic continuation to non-integer values of replicas number, $n \to 0$ \cite{29}. Nevertheless, to make a sense of replicas, one has to think to integer values of $n$. Replicas are independent equilibrium configurations sampled with the same quenched disorder. This definition corresponds to the protocol used in numerical simulation. One “replica” of the system is represented by the swarm of $N_{PT}$ configurations used for a given instance of the Parallel Tempering MC dynamics. Different instances of the PT dynamics characterized by the same set of quenched couplings $J_i$ and the same interaction network between modes are different replicas. For $N = 32$, $N = 48$ and $N = 64$ for each disorder instance we simulated 4 replicas, which gave us the availability of 6 independent values of $q^{12}$, $q^{13}$, $q^{14}$, $q^{23}$, $q^{24}$, $q^{34}$. For $N = 102$ we simulated two replicas for each instance of the disorder. To accumulate statistics for $P(q)$ we measured values of $q^{12}$ comparing replicas at the same iteration of the PT dynamics, each 500 iterations. Since the distribution $P(q)$ is not self averaging \cite{30}, for each size of the system we have sampled the equilibrium measure for $N_{sample} \approx 100$ instances of the disorder.

It is useful to clarify also how we measured in practice all thermal averages indicated with angular brackets, $\langle O[A] \rangle$. Thermal averages were measured as “time” averages along the PT dynamics, along the second half of each run:

$$\langle O[A] \rangle = \frac{2}{N_{sweep}} \sum_{i=N_{sweep}/2}^{N_{sweep}} O[A_i]. \tag{20}$$

| $N$ | $N_4$ | $T_{min}$ | $T_{max}$ | $N_{PT}$ | $N_{sweep}$ | $N_{sample}$ |
|-----|------|---------|---------|---------|------------|-------------|
| 32  | $2^{14}$ | 0.2     | 2.0     | 32      | $2^{20}$   | 100         |
| 48  | $2^{13}$ | 0.3     | 2.0     | 32      | $2^{20}$   | 100         |
| 64  | $2^{14}$ | 0.5     | 1.4     | 32      | $2^{20}$   | 100         |
| 102 | $2^{16}$ | 0.7     | 1.2     | 32      | $2^{21}$   | 100         |

**TABLE I.** Details for the simulations of NB systems for several sizes.
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