Experimental evidence of replica symmetry breaking in random lasers

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Spin-glass theory is one of the leading paradigms of complex physics and describes condensed matter, neural networks and biological systems, ultracold atoms, random photonics, and many other research fields. According to this theory, identical systems under identical conditions may reach different states and provide different values for observable quantities. This effect is known as Replica Symmetry Breaking and is revealed by the shape of the probability distribution function of an order parameter named the Parisi overlap. However, a direct experimental evidence in any field of research is still missing. Here we investigate pulse-to-pulse fluctuations in random lasers, we introduce and measure the analogue of the Parisi overlap in independent experimental realizations of the same disordered sample, and we find that the distribution function yields evidence of a transition to a glassy light phase compatible with a replica symmetry breaking.

Replica theory was originally introduced by Edwards and Anderson and detailed by Sherrington and Kirkpatrick to try to solve the prototype model for spin-glasses [1]. It was readily recognized as one of the fundamental paradigms of statistical mechanics and, after Parisi resolution of the mean-field theory [1], found applications in a huge variety of different fields of research [2, 3]. Spin-glass theory gives a rigorous settlement to the physical meaning of complexity, and describes a number of out-of-equilibrium phenomena (e.g., weak non-ergodicity and aging) [1, 4]. More recently this theory has found application in the field of random photonics [5], as specifically for random lasers (RLs) and nonlinear waves in disordered systems [6, 7]. However, notwithstanding the theoretical relevance, an experimental demonstration of the most important effect, the so-called replica symmetry breaking is still missing. Spin-glass theory predicts that the statistical distribution of an order parameter, the Parisi overlap, changes shape when a large number of competing equilibrium states emerges in the energetic landscape [8]. When this happens, replicas of the system, i.e., identical copies under the same experimental conditions, may furnish different values of observable quantities, because they settle in ergodically separated states after a long dynamics. This phenomenon is inherently different from chaos, and relies on non-trivial equilibrium properties of disordered systems.

RLs are realized in disordered media with gain; the feedback for stimulated emission of light is given by the scattering and no external cavity is needed [9]. Different RLs show multiple sub-nanometer spectral peaks above a pump threshold [10]. The wide variety of the spectral features reported and the still debated emission properties are due to the fact that RLs are open systems where light can propagate in any direction in a disordered fashion instead of oscillating between well specific boundaries as in standard lasers [11]. The scattering strength and/or pumping conditions [12–16] affect the emission: a large number of modes may result in a continuous broadband, or exhibit distinguishable resonances.

Various authors have reported evidences of shot-to-shot fluctuations in RLs. In [17] they are ascribed to the coexistence of localized and extended modes without analyzing the statistical distribution of their shot-to-shot correlations. In [18] they are due to the intrinsic matter fluctuations in fluids, that are not present in samples with static disorder as in the case here considered. In [19] the analysis of the different diffusion regimes is reported and it is not linked to a phase transition from spontaneous emission to RL, because the exponent of the diffusion law does not help to discriminate lasing from fluorescence. In [20] mode-competition and seemingly chaotic behaviour have been considered, and intended as a generic sensitivity to initial conditions, no analysis of the distribution of the shot-to-shot correlations is provided. In [21] fluctuations due to a transparent Kerr medium are theoretically considered, and even if they are not strictly related to the case with gain and loss of

RLs, they look to have connections with the spin-glass approach in [6, 7]. The latter approach has been developed in a series of papers, and is based on the mode-coupling laser theory as, for example, in [22], which reports on the time dynamics of frequency locking mechanism and without addressing the statistical properties of the laser emission. Our understanding is that the RL spectra fluctuations are a manifestation of the existence of many degenerate lasing states, each one corresponding to a given ensemble of activated modes with their own wavelengths, phases and intensities. This theory is a statistical mechanical formulation obtained starting from the decomposition in normal modes of the electromagnetic field of light, in the presence of non-linearity, and leading to a quenched disordered spin-glass-like Hamiltonian. A complex landscape typical of glassy systems is expected to occur beyond some threshold critical value of the external parameter driving the transition. The measurements reported in this manuscript confirm this theory: in a system that we believe to be properly represented by a spin-glass theory - Refs. [6, 7] - we measure the spin-glass order parameter distribution function, i.e., $P(q)$ and we find a behavior akin to the one theoretically representing the spin-glass phase (at high pumping) and the paramagnetic/fluorescence phase (at low pumping), as well as the transition between them.

RESULTS

Spin-glass theory of random lasers

In the following, we report on the shot-to-shot emission fluctuations from planar RLs made of a fluorescent π-conjugated oligomer in amorphous solid phase and we analyze the experimental results by means of the replica theory.

At variance with standard chaotic ordered lasers [23, 24], in which, for specific and tailored conditions, few modes provide exponentially diverging temporal trajectories, RLs are thermodynamic systems, with thousands of modes and degrees of freedom, which exhibits a huge number of (meta-)stable states. A standard chaotic laser will always display the same spectral behaviors in the same conditions; at large variance with what we observe in RL.

In the spin-glass approach the RL modes are treated as continuous complex spin variables with a global (power) constraint and whose coupling is governed by the interplay between disorder and nonlinearity. We remark that in this theoretical analysis, the Hamiltonian description is an effective one, representing the stationary regimes under pumping. The role of inverse temperature in equilibrium thermodynamics is played by the energy pumped into the system. The Hamiltonian description is derived in such a way to encode, in a distinct, effective temperature space, pumping and dissipation of the laser. Hence, the theoretical approach also includes the fact that RLs are open dissipative systems with an external pumping mechanism and radiation losses [6, 7]. In our experiments the disorder is fixed and the nonlinearity increases with the pumping energy, which acts as the inverse of temperature in statistical mechanics [25, 26]: at low energy (high temperature) there is no gain competition [27, 28] between the modes and they oscillate independently in a continuous wave paramagnetic regime; while at high energy (low temperature) the coexistence of mode coupling through the gain competition and frustration due to disorder gives rise to a glassy regime. In this regime a large number of electromagnetic modes is activated and in interaction. The set of the activated mode configurations is found to change from pulse-to-pulse. We will refer to each set of configurations as a state. This is justified by the fact that during a single pulse of the pump beam, very many stimulated emission processes take place at each mode frequency, that is, each mode performs a long dynamics, compatible with thermalization.

The observation of numerous different states can be understood as the evidence of a thermodynamic phase corresponding to many valleys separated by barriers in the corrugated free energy landscape. The same sample under identical experimental conditions furnishes different laser emissions. Each different instance of laser emission is, thus, a physical realization of a replica, in the meaning introduced in replica theory [1].

Random laser emission

The investigated system is a functionalized thiophene based oligomer commonly named T5OCx (see Methods for details) in amorphous solid state. RL in T5COx spin-coated in thin films [29] and lithographed in microstructures [30] has been previously observed. The strong density of the T5OCx supra-molecular laminar packing in the solid samples allows to study shot-to-shot emission fluctuations not reported so far. A sketch of the pumping and collecting geometry is given in the inset of fig. 1b. Experimental details are reported in Methods.

We illustrate in fig. 1b single shot high resolution (0.07nm) emission spectra, taken at identical experimental conditions. Input energy is 10mJ. The presence of RL modes with configuration variable from pulse-to-pulse is evident: each time the system is pumped the numerous passive modes randomly compete for the available gain, giving rise to several different compositions of the activated spectral peaks [20]. Such behavior is also evidenced by the direct visualization of the sample during pumping, as reported in fig. 2, where four different fluorescence images taken at four single shots exhibit different emission patterns and corresponding spectral features. The more intense spots signal activated modes spatial extension, changing from shot-to-shot.

In figs. 3a and 3b we show emissions of subsequent 100 shots with lower spectral resolution (0.3nm), at two different pump energies. There is no time periodicity in the spectral fluctuations: their sequence is random. At low energy the noisy variations of the spontaneous emission are negligible if compared to the pronounced fluctuations observed at high energy.

Analysis of the spectral emissions

The cavity coupled modes in the Hamiltonian describing random lasers [6] are variables not easily accessible in the experiments. We can otherwise measure spectra and have access to the intensities $I(k) \propto \langle |a_j^2| \rangle_k$, where a_j is the amplitude of the longitudinal mode j [6, 7] at wavelength λ_j. In our analysis each spectrum represents a different state of the same thermodynamic phase. The average is taken over all modes having similar wavelength, binned together in data acquisition with index k, due to the finite spectral resolution $\Delta\lambda = 0.3$ nm. We define the average spectral intensity as

$$\epsilon_\alpha = \frac{1}{N} \sum_{k=1}^{N} I_\alpha(k), \tag{1}$$

being α the replica index and N the number of acquired spectral points. The variation of this parameter from shot-to-shot quantifies the observed fluctuations. Mean and variance of ϵ_α over N_s replicas for each pumping are

$$< \epsilon >= \frac{1}{N_s} \sum_{\alpha=1}^{N_s} \epsilon_\alpha, \tag{2}$$

$$\mathrm{Var}[\epsilon] = \frac{1}{\mathrm{N_s}} \sum_{\alpha=1}^{\mathrm{N_s}} (\epsilon_\alpha - < \epsilon_\alpha >)^2, \tag{3}$$

Figure 3c shows the continuous growth for increasing input energy of the mean intensity, cf. Eq.(2). It is very important to explain the stimulated emission we are observing. In our samples we see the coexistence of RL and amplified spontaneous emission (ASE) [29, 31]. The latter is evident from the absence of threshold in the spectral peak intensities in fig. 3c and RL is evident from the fine peaks superimposed to the ASE band in fig. 1b. This specific system, made of thick amorphous solid dye, displays a threshold in terms of the spectral fluctuations, quantified by Var[ϵ], cf. Eq. (3) and fig. 3d. Such behavior has not been reported in previous works on similar dyes [29, 31]. It is well known that ASE does not exhibit a threshold, at variance with RL. However, in our case the RL threshold is masked by ASE when coming at the spectral peak. On the contrary, the threshold of the fluctuations in fig. 3d reveals the onset of the RL action, that for this sample is estimated around 3mJ. All experiments are performed in the same environmental conditions, that is the temperature, the position and the state of the sample and the performances of the optical components are controlled to be stable by growing pump energy. To check the stability of the input laser, we estimate the energy fluctuations as the percentage variation given by the ratio between the standard deviation and the mean value of the emission peak intensity with and without sample, for increasing energy. The plots are reported in the inset of fig. 3d, where the laser energy variations are approximately around 4% at any energy, while the same quantity from the sample increases at high energy. We stress that an error of 4% on the x-axis of fig. 3d is inside the symbols and negligible compared to the energy values, thus it cannot cause the evident increase of the observed variance.

To analyze the system behavior in the framework of statistical mechanics of disordered systems, and characterize the high pumping RL regime, we introduce another order parameter to identify the phase transition: the overlap between intensity fluctuations in different experimental replicas. The experimentally accessible variable, coarse graining the behavior of single modes, is the intensity fluctuation at a given frequency:

$$\Delta_\alpha(k) = I_\alpha(k) - \bar{I}(k) \tag{4}$$

with $\bar{I}(k)$ the average over replicas of each mode intensity

$$\bar{I}(k) = \frac{1}{N_s} \sum_{\alpha=1}^{N_s} I_\alpha(k). \tag{5}$$

The overlap between pulse-to-pulse intensity fluctuations is defined as

$$q_{\alpha\beta} = \frac{\sum_{k=1}^{N} \Delta_\alpha(k) \Delta_\beta(k)}{\sqrt{\sum_{k=1}^{N} \Delta_\alpha^2(k)} \sqrt{\sum_{k=1}^{N} \Delta_\beta^2(k)}} \tag{6}$$

From the measured spectra we calculate the set of all $N_s(N_s-1)/2$ values q of $q_{\alpha\beta}$ for each different input energy, determining their distribution $P(q)$. We retain $P(q)$ as a measure of the theoretical distribution of the overlap between mode amplitudes [6, 7], which is a leading quantity for the description of glassy phases [1]. So far no experimental measurements of $P(q)$ have been reported.

Six examples are reported in fig. 4a-f for increasing pump energy. At low energy (fig. 4a) all overlaps are centered around the zero value, meaning that the electromagnetic modes are independent and not interacting in the paramagnetic regime. By increasing energy, modes are coupled by the nonlinearity and this corresponds to a non-trivial overlap distribution (fig. 4b-f). In the high energy glassy phase, with all modes highly interacting and frustrated by the disorder, q assumes all possible values in the range [-1,1]. Such behavior of the $P(q)$ evidences the fact that the correlation between intensity fluctuations in any two replicas depends on the replicas selected. The variety of possible correlations extends to the whole range of values. This is a manifestation of the breaking of the replica symmetry.

In fig. 4g we show $q_{\max}$ corresponding to the position of the maximum of $P(|q|)$ versus pumping: it changes drastically signaling a phase transition between 2mJ and 3mJ, compatible with the threshold determined by $\mathrm{Var}[\epsilon]$ in fig. 3d. We stress that in the high pump regime $P(q) > 0$ for any value of q.

DISCUSSION

We experimentally show that the RL intensity fluctuations grow drastically as the pump energy increases beyond a threshold, exhibiting a transition from negligible to large oscillations. A possible order parameter is quantified by the variance of the fluctuations. A more refined one is the distribution of all mutual overlaps between intensity fluctuations in different physical replicas. Besides evidencing the threshold of phase transition, it describes the organization of states and their non-trivial correlation, non-symmetric in replicas exchange. Above the threshold pump energy, the overlap between two replicas does depend on the specific couple chosen and the variation from couple to couple turns out to be more complicated than the ordinary statistical fluctuations. That is, replicas are not all equivalent to each other and we, thus, observe an instance of replica symmetry breaking through the random laser transition.

The parameter q measures the overlap of the shot-to-shot fluctuations at each wavelength around the average spectrum at a given pumping. Thus $q \sim 1$ when two shots both have very similar intensity fluctuations at each wavelength. In principle, it does not matter how large the fluctuations are to have a large q, but only how much they are correlated. The fact that large overlap values become probable in the lasing regime above threshold is not a trivial feature. Similarly, one has $q \sim -1$ when two shots display spectral fluctuations similar in magnitude (no matter how large) but opposite in sign, that is the shot-to-shot fluctuations around the average spectrum are very anti-correlated. When $q \sim 0$ the intensity fluctuations in two shots are basically uncorrelated. This can imply the ASE background, but can also correspond to the presence of a sub-set of shots with uncorrelated large fluctuations. Above threshold, the latter situation occurs, as can be observed looking at the single spectra. By the definition of overlap in Eq. (6) in all cases $|q| \sim [0, 1]$, the fluctuations of a single spectrum can be large or small. The important observation is that increasing the pumping, the correlations between shot-to-shot fluctuations become non-trivial right when the fluctuation magnitude starts increasing significantly (i.e., above threshold), cf. fig. 3d.

As a last remark, we recall that in the effective statistical mechanical Hamiltonian for random lasers the variables are, actually, the modes, i.e., complex amplitudes [6, 7], not easily accessible in the experiments. Whereas we are considering their squared modulus given by the emitted intensity, thus losing in formation about the actual state structure. Looking at the present results we stress, however, that such coarse-graining appears to be refined enough to investigate the peculiar properties of glassy-like regimes of random lasers.

METHODS

Sample preparation and characterization. The investigated disordered system is a functionalized thienyl-S,S-dioxide quinquethiophene (T5OCx), whose preparation modalities and molecular structure are reported elsewhere [32], in amorphous solid state obtained by the crystallization of the dye powder after a thermal shock above the transition temperature (T_g). Successively, the packing states have been frozen by cooling across the T_g. The thermal process confers to the π-conjugated oligomer a crystal-like molecular ordering over large area with the appearance of laminar domains that grow anisotropically in time up to their final random packing leading, at the end of the process, to a macroscopic disordered system as shown in fig. 1a. Sample characterization is performed by a z-stack sequential acquisition of confocal microscope (Olympus, FV1000). The optical sections are collected in transverse xz- and yz-planes. The x-y surface area of the investigated samples is in the range (10^5-10^6)μm^2 and the average thickness along z is (7±2)μm.

Measurements. The samples are pumped by a frequency doubled Q-switched Nd:YAG pulsed laser emitting at λ=532 nm, with 10 Hz repetition rate, 6 ns pulse duration and with 12 mm beam diameter, the emitted radiation is collected from the sample edge into an optic fiber connected to a spectrograph equipped with electrically cooled CCD array detector, with gratings density of 600 mm^{-1} and 1800 mm^{-1}, for low (0.07nm) and high (0.3nm) resolution spectra respectively. The samples are put on glass slides and pumped perpendicularly along the z direction and all reported spectra are taken from single pumping shots.

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STATEMENT OF AUTHORS CONTRIBUTION

N. G. and C. C. worked on the experimental characterization of the random laser emission. I. V., F. D.M., G. B. and G. G. worked on the samples preparation and characterization. L. L. worked on the theoretical interpretation of the data. N. G., L.L., and C.C. wrote the manuscript.

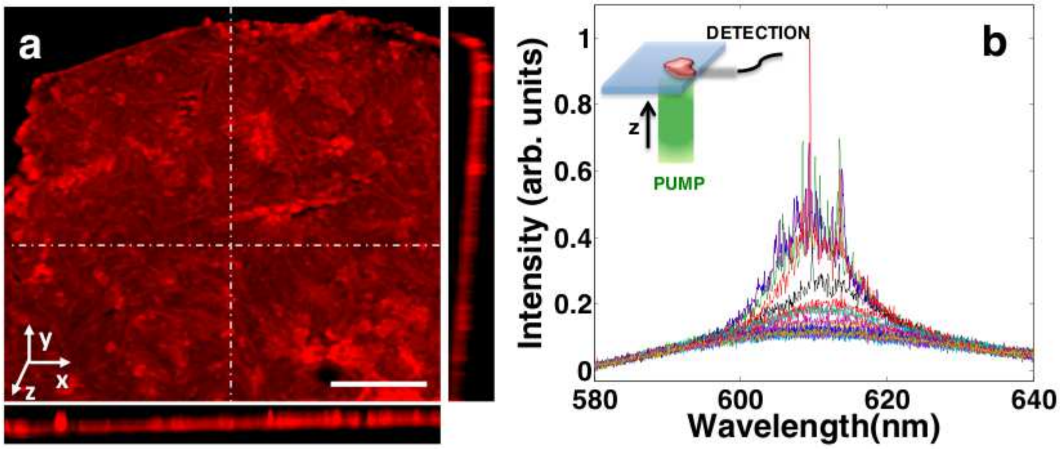

FIG. 1. **Sample image and emission spectra showing random lasing.** (a) 3D-reconstruction of confocal microscopy Z-stack images of the supra-molecular laminar packing in aT5OCx solid sample. The right and the bottom panels report the yz- and the xz-sections, respectively. Scale bar: 20μm. (b) High resolution single shot spectra taken in the same conditions, 10mJ pump energy. Inset: sketch of the experiment.

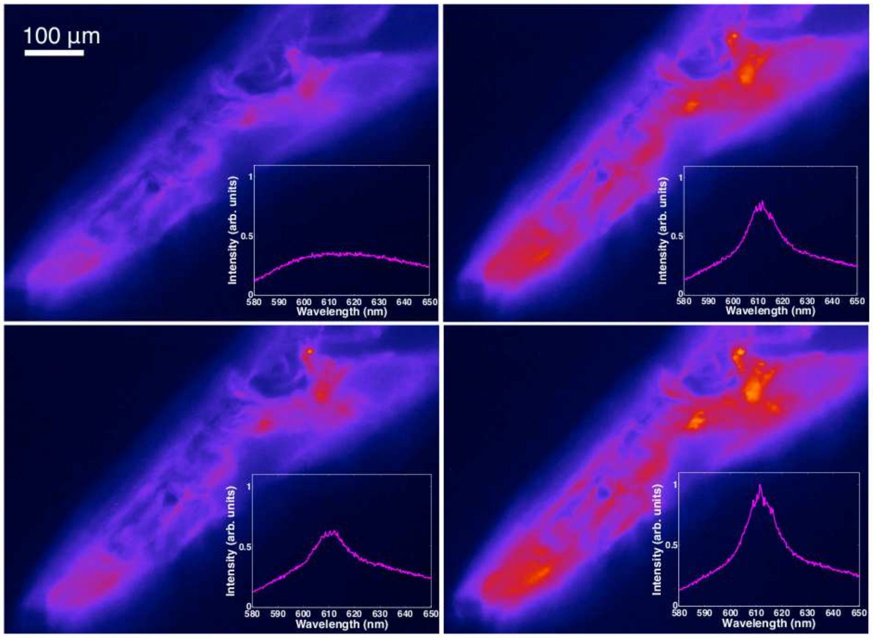

FIG. 2. **Snapshots of RL emissions.** Single shot optical images and corresponding emission spectra (insets) during the pumping of the sample in the same experimental conditions. The input energy is 10mJ.

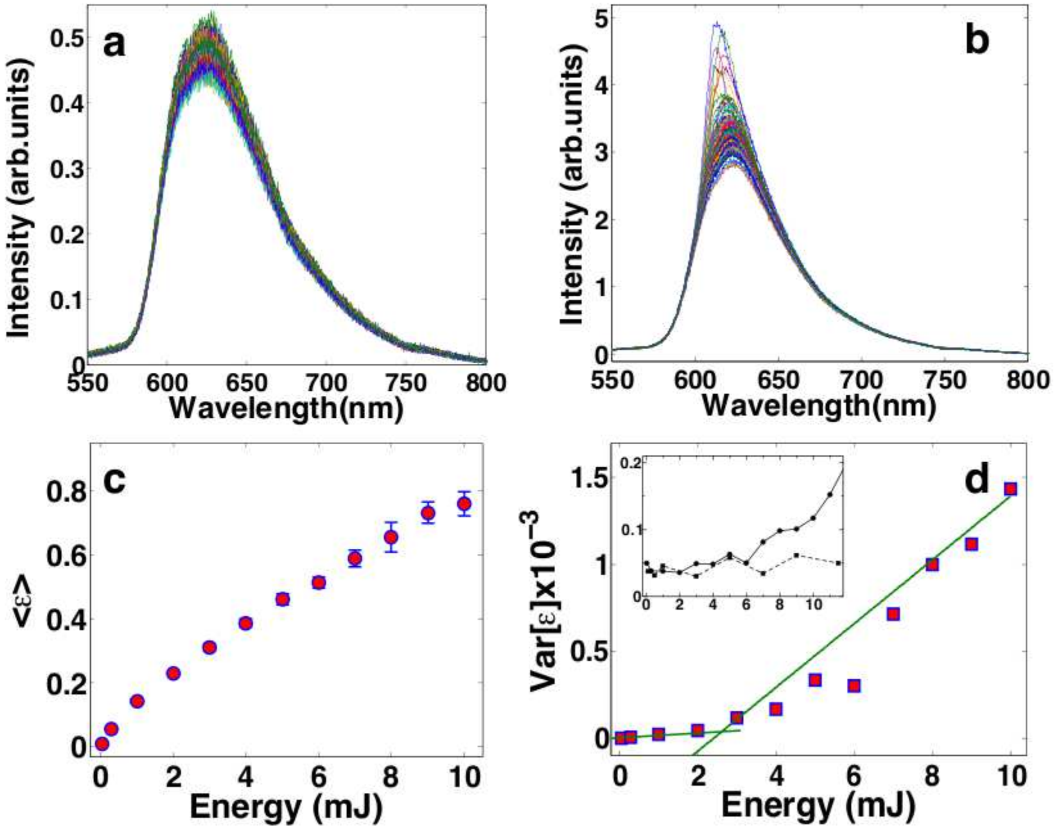

FIG. 3. **Fluctuations in the random laser emission.** a-b) Emission spectra at low energy 1mJ (a) and high energy 12mJ (b). c) Mean emission intensity $< \epsilon >$ vs. pump energy, error bars are standard deviation. d) Variance of ϵ vs. pump energy. Inset: peak standard deviation divided by mean peak intensity from the sample (solid line) and the pump laser (dashed line) emission spectra.

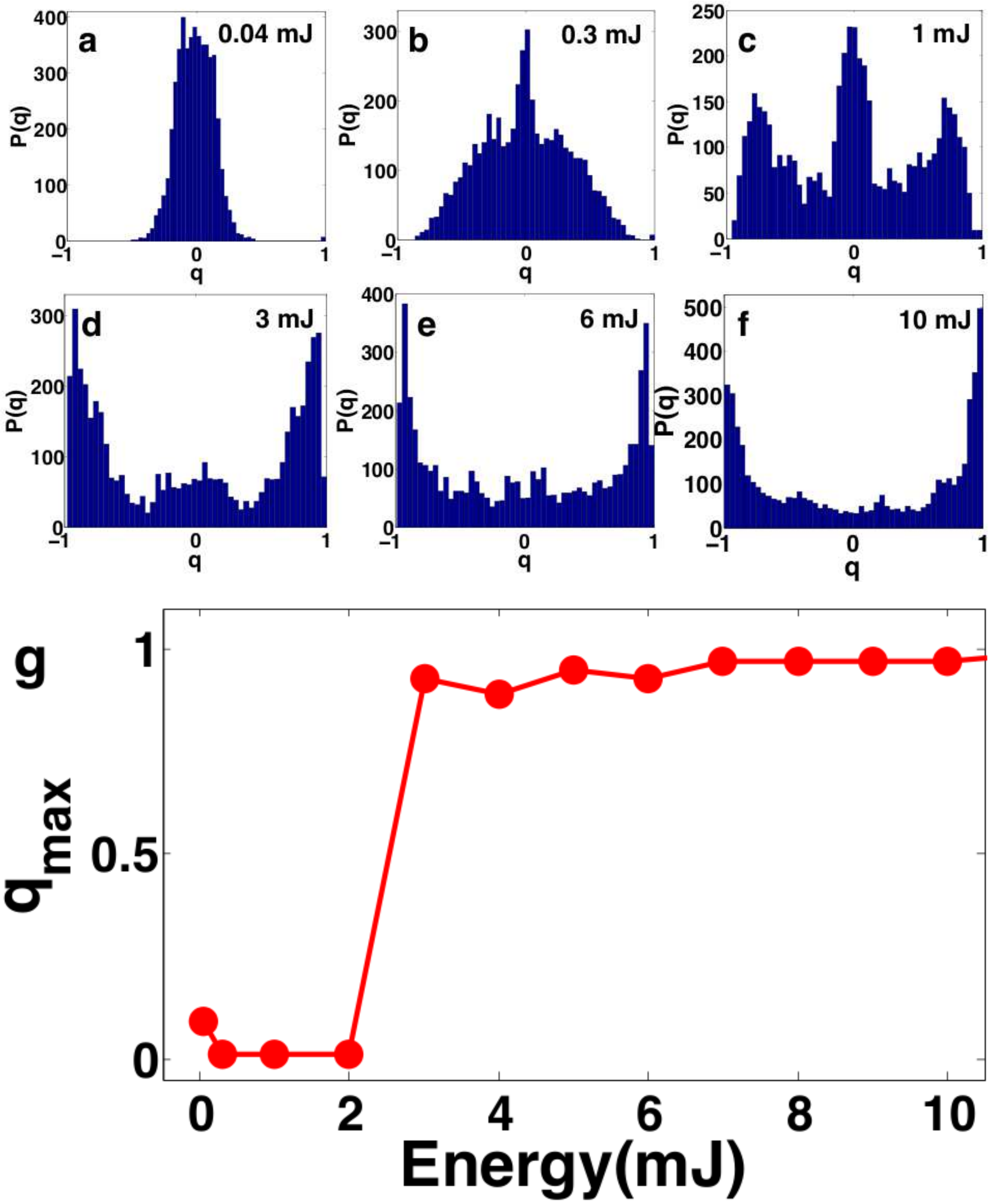

FIG. 4. **Distribution function of the overlap showing replica symmetry breaking by increasing pump energy.** a-f) Distribution of the overlap q at different pump energy. g) $q_{\max}$ corresponding to the position of the maximum of $P(|q|)$ versus pumping.