Replica Symmetry Breaking in the Photonic Ferromagneticlike Spontaneous Mode-Locking Phase of a Multimode Nd:YAG Laser

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We demonstrate the replica symmetry breaking (RSB) phenomenon in the spontaneous mode-locking regime of a multimode Q-switched Nd:YAG laser. The underlying mechanism is quite distinct from that of the RSB recently observed in random lasers. Here, there is no random medium and the phase is not glassy with incoherently oscillating modes as in random lasers. Instead, in each pulse a specific subset of longitudinal modes are activated in a nondeterministic way, whose coherent oscillation dominates and frustrates the others. The emergence of RSB coincides with the onset of ultrashort pulse generation typical of the spontaneous mode-locking regime, both occurring at the laser threshold. On the other hand, when high losses are introduced, RSB is inhibited and only the amplified stimulated emission with replica symmetry is observed. Our results disclose the only theoretically predicted photonic phase with RSB that remained unobserved so far.

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The recent reports of the replica symmetry breaking (RSB) phenomenon in photonic experiments boosted the understanding of the interplay of disorder or randomness, amplification, and nonlinearity in multimode lasers [1–5]. They also helped to settle quite enlightening connections between photonics and the statistical physics of complex systems [6–11]. RSB manifests itself when identically prepared system replicas reach distinct states, yielding different measures of observable quantities [12]. The concept of RSB appeared in the context of Parisi's theory of disordered magnetic systems [12]. In that framework, for sufficiently low temperatures, the free energy landscape breaks into a large number of local minima in the configuration space. As a given configuration can be trapped for a long time in a local minimum, replicas become no longer equivalent, leading to distinct values of observables and nontrivial correlation patterns in the RSB scenario.

The prototype of a magnetic system that exhibits RSB is the spin glass [12]. In a glassy state, the spins fail to align in a spatially regular configuration due to strongly frustrated magnetic interactions. Instead, they "freeze" along random directions at low temperatures. Remarkably, RSB can also arise in a ferromagnet with random spin bonds [13]. In this case, the low-temperature states present frustrated spin domains nucleated in a predominantly ferromagnetic background of aligned spins.

In the photonic context, theoretical predictions of RSB emerged in the last decade [6–11]. In the photonic-to-magnetic analogy, the amplitudes of the active optical modes play the role of the spins, and the excitation pumping power acts as the inverse temperature. Two rather distinct photonic phases were predicted to display the RSB phenomenon [6,9,10]. On the one hand, the photonic spin-glass phase with RSB has already been experimentally demonstrated in solid-state [1,2] and colloidal [3] random lasers, as well as in a random fiber laser [4,5]. All these cavityless random lasers present some type of strong embedded disorder (e.g., randomly placed scatterers) that causes the modes to be nontrivially correlated, while displaying incoherent oscillation (in a way analogous to the frustrated spins frozen at random directions in a spin-glass phase).

On the other hand, the theoretical analysis also predicts [6,9,10] the remarkable possibility of RSB with weak disorder or randomness in a mode-locking laser regime. The magnetic analogue of the RSB mode-locked lasing phase, with most active modes oscillating coherently, is the RSB random-bond ferromagnetic system, with mostly aligned spins dominating over small frustrated spin clusters. However, different from the photonic RSB spin-glass phase, the RSB mode-locking properties have not been experimentally observed so far.

Regarding the nature of the disorder and frustration underlying the photonic RSB, a recent work [14] demonstrated that solid-state and liquid multimode lasers without any form of intentionally added disorder can also present RSB in the threshold region with strong intensity fluctuations. In that case, the large laser cavity allowed for many modes, whose simultaneous coherent oscillation was

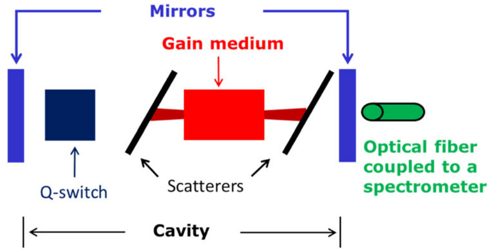

FIG. 1. Q-switched Nd:YAG pulsed laser with single-pulse spectra collected by a multimode optical fiber coupled to a spectrometer. A large cavity of effective length 0.60 m gives rise to the multimode character. If high losses are introduced by placing efficient scatterers between the crystal and the cavity mirrors, the laser oscillation is suppressed.

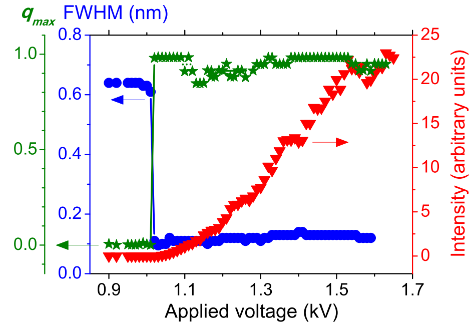

FIG. 2. Total emitted intensity (red triangles), averaged over 500 spectra for each applied voltage, FWHM (blue circles), and the parameter $q_{\max}$ (green stars) at which the distribution $P(q)$ of replica overlaps of pulse-to-pulse intensity fluctuations is maximum as a function of the applied voltage V_{app}. The abrupt change in FWHM at the threshold from the ASE to the SML laser regime remarkably coincides with the sharp transition from the replica-symmetric ($q_{\max} \sim 0$) to the RSB ($q_{\max} \sim 1$) regime.

hindered, leading to photonic frustration. In the absence of the usual structural disorder, randomness was intrinsically related to the setting of the dominating modes. However, we remark that the reported RSB phase in [14] did not show mode-locking properties.

In the present Letter, we demonstrate the RSB phenomenon in the spontaneous mode-locking (SML) laser regime of a multimode Q-switched Nd:YAG laser. In contrast with the random lasers with RSB spin-glass behavior, there is no random medium with embedded disorder, and the photonic RSB phase is not glassy with incoherently oscillating modes. Here, in each pulse of the pump source a specific subset of longitudinal modes are activated in a nondeterministic way, whose ferromagnetic-like coherent oscillation dominates and frustrates the other modes. The evidence of RSB is attested by the calculation of statistical correlations among the intensity fluctuations. As a further and independent experimental verification of the SML regime, we employed an ultrafast temporal measurement system to analyze the time-domain behavior of the ultrashort pulses and found that their emergence remarkably coincides with the onset of the RSB properties. It is worthwhile noting that SML in lasers is not a novel subject, and has been theoretically and experimentally studied in several works [15–19], in the context of ultrashort laser pulse generation. However, the connection with statistical physics based models has only been pointed out in [6,9,10] and demonstrated here for the first time. Last, we further report that the laser action is actually essential to the occurrence of RSB. Indeed, when the laser oscillation is suppressed by introducing high losses in the cavity, the RSB behavior is inhibited and only the amplified stimulated emission (ASE) with replica symmetry is observed.

The experimental setup, shown in Fig. 1, consisted of a Nd:YAG pulsed laser system (5 Hz repetition rate, 7 ns nominal pulse duration), driven by a Q-switcher device. The excitation pump source was a xenon flash lamp. Single-pulse spectra were collected by a multimode optical fiber coupled to a spectrometer with nominal resolution of 0.024 nm. For each voltage V_{app} applied to the flash lamp, data from 500 spectra were recorded and analyzed. The multimode character of the laser system arises from the large cavity of length 0.50 m (effective length of 0.60 m when the refractive indices of the intracavity elements are considered). This setup allows the emergence of mode competition from the significant spatial superposition, known to be a relevant mechanism in such laser systems [20]. On the other hand, mode-locked lasers with saturable absorbers are much distinct from the present one, as discussed in [9,21].

In Fig. 2, the total emitted intensity and linewidth reduction, inferred by the full width at half maximum (FWHM), are plotted as a function of V_{app}. Averages were taken over 500 spectra. The abrupt change in FWHM (blue circles) indicates the laser threshold voltage value of 1.01 kV, which coincides with the prelasing ASE to the SML laser transition regime (see, also, below for an independent verification of the SML regime). As V_{app} is increased, the total emitted intensity (red triangles) enhances smoothly.

The identification of the RSB properties in a photonic system relies [6–11] on the analysis of mode-mode correlations among replicas, which can be calculated by the replica overlap parameter [1,6],

$$q_{\gamma\beta} = \frac{\sum_k \Delta_\gamma(k)\Delta_\beta(k)}{\sqrt{\sum_k \Delta_\gamma^2(k)}\sqrt{\sum_k \Delta_\beta^2(k)}}, \tag{1}$$

where $\gamma, \beta = 1, 2, \ldots, N_s$ denote the replica labels ($N_s = 500$ in the present work), the mean intensity at the wavelength indexed by k is $\bar{I}(k) = \sum_{\gamma=1}^{N_s} I_\gamma(k)/N_s$, and the intensity fluctuation is given by $\Delta_\gamma(k) = I_\gamma(k) - \bar{I}(k)$. In the photonic context, each intensity spectrum generated by a laser pulse is considered a replica, i.e., a copy of the system under fairly identical experimental conditions (in fact, this assumption is properly examined below).

The probability density function (PDF) $P(q)$, analogous to the Parisi order parameter in replica theory [12], describes the distribution of replica overlaps $q = q_{\gamma\beta}$, signaling a replica-symmetric or a RSB phase if it peaks only at $q_{\max} = 0$ or, also, at absolute values $q_{\max} \neq 0$, respectively.

We show the PDFs $P(q)$ in Fig. 3, along with the respective profiles of a fraction (20) of the 500 recorded spectra for each V_{app}. The indicated voltages are representative of the prelasing (0.90 kV), laser threshold (1.01 kV), right above threshold (1.02 kV), and well developed SML laser (1.63 kV) regimes. In the former two cases, the distributions display a single peak at $q_{\max} = 0$ [Figs. 3(a) and 3(b)], which evidences a replica-symmetric phase. This regime corresponds to the prelasing ASE behavior with weak pulse-to-pulse intensity fluctuations [Figs. 3(e) and 3(f)]. Right above the threshold, the PDF $P(q)$ starts to present a double-peaked pattern, with $q_{\max} \neq 0$ [Figs. 3(c) and 3(d)]. This result signalizes the onset of the RSB phase at the threshold to the SML behavior. In this regime, strong fluctuations of the maximum intensity are observed in Figs. 3(g) and 3(h), and the laser action takes place with the narrow linewidth.

The transition from the replica-symmetric to the RSB phase can be identified in the plot of $q_{\max}$ against V_{app}

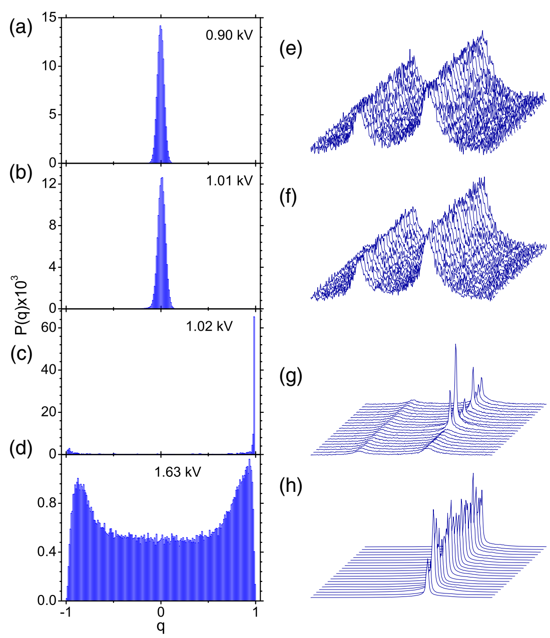

FIG. 3. (a)–(d) PDFs $P(q)$ of replica overlaps of pulse-to-pulse intensity fluctuations in 500 emission spectra, for applied voltages in the prelasing ASE (0.90 kV), threshold (1.01 kV), nearly above threshold (1.02 kV), and SML laser (1.63 kV) regimes. Replica-symmetric and RSB behaviors are, respectively, related to distributions displaying a single peak at $q_{\max} = 0$ and double peaks with $q_{\max} \neq 0$. (e)–(h) 20 emission spectra at each V_{app} associated with the PDFs shown in (a)–(d). Fluctuations of the maximum intensity become rather strong in the RSB SML laser regime.

(Fig. 2). We highlight the remarkable agreement between the abrupt change in FWHM (blue circles), at the threshold from the ASE to the lasing regime, and the sharp transition from the replica-symmetric ($q_{\max} \sim 0$) to the RSB ($q_{\max} \sim 1$) regime (green stars). SML is clearly observed above the threshold (see, also, below).

At this point, a careful investigation of the above assumption of the establishment of system replicas after each excitation pulse of the flash lamp is in order. First, to verify that the presented results are not a spurious artifact of the flash-lamp fluctuations, we analyzed 500 spectra emitted by the flash lamp for each one of the regimes of Fig. 3, just as was done for the Nd:YAG laser device. As shown in Fig. 1S of the Supplemental Material [22], at any given voltage the flash-lamp spectra are, indeed, rather similar. Moreover, their fluctuation patterns also remain essentially unaltered as the voltage is varied. Thus, the drastic change of behavior noted in Figs. 2 and 3 as the threshold is crossed cannot be attributed to the flash-lamp properties. These findings also corroborate the assumption that, after each pulse at a given voltage, the system is subjected to fairly identical experimental conditions, which, nevertheless, can result in different measures of observables, such as the emission spectra above threshold [see Figs. 3(g) and 3(h)]. Such a description is fully compatible with the concept of system replicas introduced in the replica theory [12].

Further, we mention that the peak intensity fluctuations of the flash lamp are very small, for all voltages and wavelengths within the appropriate excitation bands (see Fig. 2S of the Supplemental Material [22]). Indeed, the standard deviation (SD) of the flash-lamp peak intensity fluctuations displays a nearly constant value of less than 2% independently of the voltage, contrasting with the increasing SD behavior of the laser system as V_{app} is enhanced above threshold.

As an independent verification of the SML behavior, we performed a high-resolution time-domain analysis of the laser pulses emitted by the Nd:YAG laser device. Using a fast photodetector (10 GHz) combined with a 20 GHz bandwidth sampling scope, we measured the temporal characteristics of the emitted radiation as a function of the applied voltage in the regimes below, near, and above the threshold. The complete results are presented in the Supplemental Material [22].

First, we comment that different regimes are, indeed, characterized through this temporal analysis, in agreement with the above discussion. Below the threshold at 0.98 kV, the FWHM pulse width of the envelope is 125.7 ns, a value already much shorter than the lifetime of the Nd:YAG level (230 μs). As V_{app} crosses the threshold value of 1.01 kV, the envelope narrows considerably [see Fig. 3S(a) of the Supplemental Material [22]]. For example, the envelope narrowing of 71% right above the threshold, from 1.01 to 1.04 kV, is much more pronounced than the 13% narrowing measured right below, from 0.98 to 1.01 kV, indicating that a rather distinct laser regime establishes at the threshold.

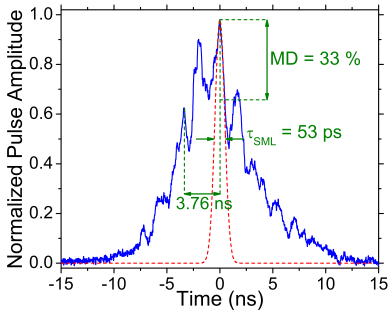

FIG. 4. Time-domain profile of a pulse train (solid blue line) emitted by the Nd:YAG laser for the applied voltage 1.30 kV (above threshold). The 3.76 ns repetition rate implies a cavity length of 0.56 m in good agreement with the effective value of 0.60 m. The pulse structure resembles more a Q-switched and mode-locked pulse train than just a mode-locked one. The dashed red line depicts the estimate of a full mode-locking pulse with maximum duration $\tau_{\mathrm{SML}} = 53$ ps, inferred from the 33% modulation depth (MD). The overall pulse envelope FWHM is 6.1 ns.

Remarkably, this finding also coincides with the onset of the RSB properties as independently determined above.

As the voltage is increased further, the envelope pulse width reaches around a 6.1 ns wide well above threshold at 1.30 kV (see Fig. 4). This result compares well with the nominal value of 7 ns for the Q-switched output duration furnished by the laser fabricant. In particular, in this regime the analysis of Fig. 4 reveals that the emitted pulse (solid blue line) resembles more a Q-switched and mode-locked pulse train, with a pulse repetition rate of 266 MHz within the envelope. For the sake of clarity, here we keep the SML notation. The pulses in this case are not fully developed to have 100% modulation depth, but from the 33% modulation depth, we can infer that the pulse structure in the Q-switched envelope corresponds to the mode-locked pulses from which a maximum pulse duration of $\tau_{\mathrm{SML}} = 53$ ps is estimated (dashed red line). We also note that the pulse repetition rate indicated above corresponds to a temporal separation among pulses of 3.76 ns, which is related to the round-trip time in a cavity of length 0.56 m in good agreement with the effective cavity length of 0.60 m.

One should also notice in Fig. 4 that, besides the marked peaks with the cavity round-trip time, another peak appears in between. The origin of this second pulse is not clear since it is not related to two pulses per cavity round trip due to their asymmetrical position. Multiple cavity pulses (per round trip) have been observed before in mode-locked lasers [17,18], but their pulse evolution is different from that observed here, as discussed in the Supplemental Material [22]. Only the peaks that survive increasing flashlamp voltage are used to compute the cavity round-trip time.

The above conclusions are strengthened by the experiment in which high losses are introduced in the cavity of the very same Nd:YAG laser by placing two pieces of paper playing the role of efficient scatterers, indicated in Fig. 1. As laser oscillations are now suppressed, the SML laser regime cannot establish, and the ASE behavior with replica symmetry is observed for all voltages, as shown in Figs. 4S and 5S of the Supplemental Material [22].

We now turn to the discussion on the mechanism underlying the RSB behavior in the SML laser regime of the multimode Q-switched Nd:YAG laser.

As mentioned, after each excitation pulse of the flash lamp a specific set of spatially overlapped longitudinal modes are activated in a nondeterministic way. Since the modes intrinsically compete for the gain, photonic frustration is induced in the sense that the coherent oscillation of a given subset of coupled modes dominates and inhibits the coherent oscillation of the others. The set of dominating modes changes stochastically from pulse to pulse. Each associated spectrum emitted under identical experimental conditions is considered a photonic replica, as discussed. This feature allows the emergence of strong pulse-to-pulse intensity fluctuations above threshold, consistently with the RSB scenario. In the statistical physics language, each photonic replica occupies a distinct thermodynamic state, and the characterization of the intensity fluctuations reveals the emergence of RSB behavior in the SML laser regime.

Finally, we stress the important contrast between our findings and the recent observation of RSB in the spin-glass phase of random lasers [1–5]. Indeed, the photonic phases are quite distinct. By sticking to the magnetic-to-photonic analogy, most active modes oscillate coherently in the ferromagnetic-like SML laser regime of the multimode Nd:YAG laser, whereas in the spin-glass-like phase of random laser systems, they present incoherent oscillation [6–11]. In this sense, our work represents the first experimental demonstration of the photonic SML laser regime with RSB theoretically predicted in [6,9,10].

In this context, we also recall that the SML laser regime was observed [23] in a random laser consisting of micrometer-sized clusters of titania nanoparticles in a Rhodamine dye solution without active or passive device. However, the occurrence of RSB was not studied in [23], despite the theoretical predictions. We also comment that a 30 ps pulsed laser was used as the pump source, and the signature of mode locking was characterized by the time-resolved analysis of the temporal emission in the nanosecond regime, similar to the present work. In [23], a 30% pulse width reduction was reported.

In conclusion, in this work a multimode Nd:YAG laser was employed to unveil the RSB behavior in the SML laser phase. The SML regime, in fact a Q-switched and mode-locking regime, was also independently demonstrated through high-resolution temporal measurements. The signature of RSB was clearly identified in the distribution of replica overlaps of pulse-to-pulse intensity fluctuations, and the underlying photonic mechanism was discussed. Moreover, the introduction of high losses in the cavity to suppress the laser action led to the observation of the ASE regime with replica symmetry. Our results disclose the only theoretically predicted photonic phase with RSB that remained unobserved so far.

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