



# Order transfer in a hybrid Raman-laser-optomechanical resonator

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**Abstract:** Order is one of the most important concepts to interpret various phenomena such as the emergence of turbulence and the life-evolution process. The generation of laser can also be treated as an ordering process in which the interaction between the laser beam and the gain medium leads to the correlation between photons in the output optical field. Here, we demonstrate experimentally in a hybrid Raman-laser-optomechanical system that an ordered Raman laser can be generated from an entropy-absorption process by a chaotic optomechanical resonator. When the optomechanical resonator is chaotic or disordered enough, the Raman-laser field is in an ordered lasing mode. This can be interpreted by the entropy transfer from the Raman-laser mode to the chaotic motion mediated by optomechanics. Different order parameters, such as the box-counting dimension, the maximal Lyapunov exponent, and the Kolmogorov entropy, are introduced to quantitatively analyze this entropy transfer process, by which we can observe the order transfer between the Raman-laser mode and the optomechanical resonator. Our study presents a new mechanism of laser generation and opens up new dimensions of research such as the modulation of laser by optomechanics.

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## 1. Introduction

Order is a quantity commonly describing particular states with randomness or uncertainty. Order exists in many systems and can be evaluated by entropy in the second law of thermodynamics. In the second law of thermodynamics, the change of the system's entropy is induced by the information flow between the system and its environment. In a broader case, the change of the degrees of order is induced by the exchange of matter and energy between the system and its environment. This exchange of matter and energy leads to the so-called generalized second law of thermodynamics [1]. According to the second law of thermodynamics, the entropy of any isolated system always increases, which means that the isolated system spontaneously changes from an ordered state to a disordered state [2]. However, the opposite process, i.e., the entropy-decreasing process or the so-called ordering process, is more interesting. It often leads to various meaningful phenomena, e.g., the evolution of life and species. The stationary states of the ordering processes are called dissipative structures. To realize a dissipative structure, the system should be open,

i.e., to exchange entropy, matter, or energy with the outside world continuously, and far away from the equilibrium state. When changes in the external conditions achieve certain threshold values, the stable and ordered dissipative structures can be spontaneously generated by the role of the internal self-organization phenomena [3]. These ordering processes and the generation of dissipative structures can be seen in many systems, such as quantum systems [4–6] and biological systems [7,8]. In addition, many new ordered structures can be realized in different materials such as metals [9,10], antiferromagnets [11], glasses [12,13], and polymers [14,15].

A laser can also be generated by an ordering process [16–21]. When the photon with a fixed frequency outcompetes the action of other photons, it is strengthened and occupies a dominant position. The process of ordering in the laser is completed when the pump power achieves a threshold value and the individual photons behave in a fully correlated manner. The ordered states of lasers can also be treated as dissipative structures [22]. As a special kind of laser, the Raman laser is realized by scattering processes caused by the vibrations of molecules, and it can be widely used in various aspects such as creating a cascaded laser source [23]. With the development of related technologies, nonlinear Raman source in optical microresonators and enhanced Raman scattering has been presented [24,25]. Raman lasers have also been successfully demonstrated in on-chip microresonators under on-demand control [26] and used for ordering process demonstration [27] recently.

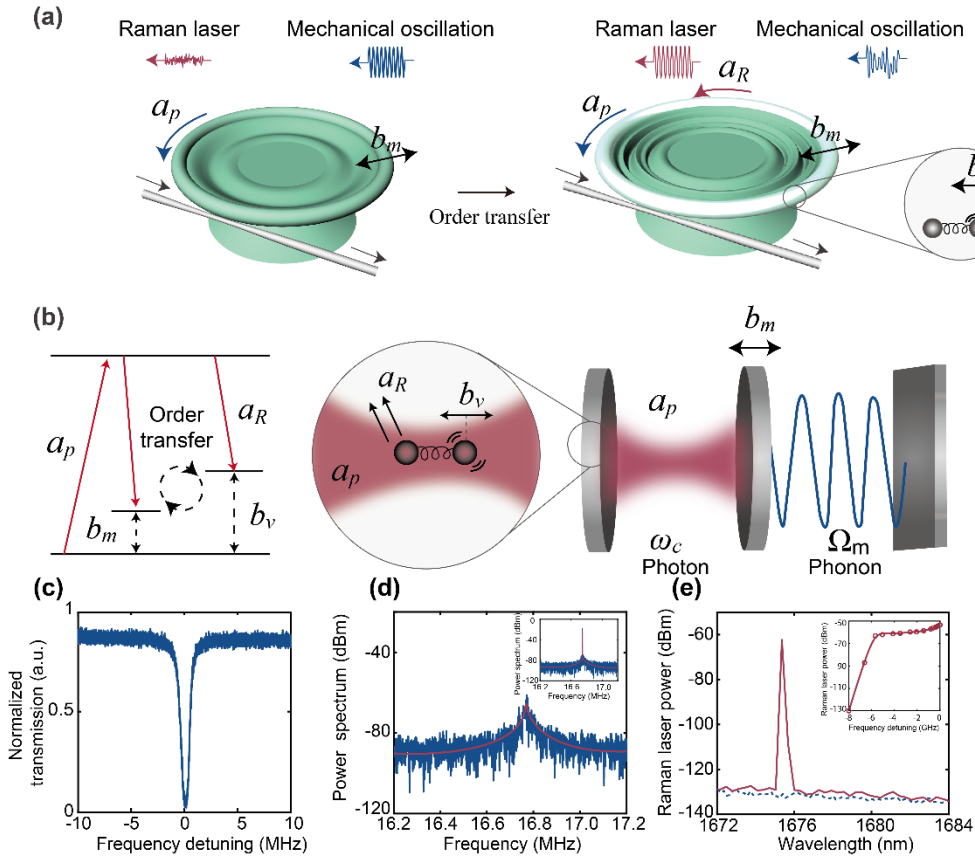
Optomechanics is another important phenomenon that can be achieved in microresonators [28]. It has been used for quantum information processing [29], detection of gravitational waves [30,31], and optical-to-microwave conversion [32]. Disordered chaotic vibrations can also be excited in optomechanical resonators with increasing optical input powers [33–36]. However, the ordered laser effects and disordered chaotic motions are somewhat conflicted so that they are hardly observed simultaneously.

In this work, we report the experiments that ordered Raman laser and disordered chaos can be shown simultaneously in a hybrid Raman-laser-optomechanical system. In this hybrid system, four modes are involved: an optical pump mode in the 1550 nm band, an optical Raman-laser mode in the 1676 nm band, a mechanical mode, and a molecular vibration mode. It has been demonstrated that the Raman laser is enhanced in this hybrid system when the optomechanical resonator turns to be more chaotic and disordered. This can be explained by the entropy-decreasing at the Raman-laser degree absorbed by an external disordered source, making the Raman-laser degree more ordered and finally stabilized at a localized state. Different order parameters such as the box-counting dimension, the maximal Lyapunov exponent, and the Kolmogorov entropy are introduced to quantitatively analyze this entropy-transfer process. Furthermore, the phase transition and the bifurcation diagram of the hybrid system during the formation of the dissipative structures are also analyzed.

## 2. Characterization of the hybrid Raman-laser-optomechanical resonator

The schematic diagram of our hybrid Raman-laser-optomechanical resonator is shown in Fig. 1(a). A tunable pump laser in the 1550 nm wavelength band is used to generate a Raman laser in a silica microtoroid with the major diameter of about 50  $\mu\text{m}$ . A fiber taper is used to couple the pump light into the microtoroid resonator. When the mechanical motion of the microtoroidal optomechanical resonator transits from the periodic regime to the chaotic regime, the vibrations of silica molecules are excited, resulting in the generation of an ordered Raman laser.

The system we consider here is a hybrid Raman-laser-optomechanical resonator, which is composed of four modes: an optical pump mode with complex amplitude  $a_p$ , a Raman-laser mode with complex amplitude  $a_R$  and eigenfrequency  $\omega_c$ , the mechanical mode with complex amplitude  $b_m$  and frequency  $\Omega_m$ , and the molecular vibration mode with complex amplitude  $b_v$  and frequency  $\Omega_v$  [Fig. 1(b)]. The optical cavity modes are coupled to the mechanical mode via the radiation forces, which can be seen as an equivalent Fabry-Perot cavity with one moving



**Fig. 1.** The hybrid Raman-laser-optomechanical resonator. (a) Cross section of the Raman-laser-optomechanical microtoroid resonator showing the indirect interaction mediated by the optical pump mode between the mechanical motion of the microtoroid resonator and the Raman-laser mode. (b) Schematic diagram of the indirect coupling between the Raman-laser mode and the mechanical vibration of the cavity mediated by the optical pump mode in a Fabry-Perot cavity with one moving end mirror. (c) The normalized transmission of the optical pump mode in the 1550 nm band with a quality factor of  $4.2 \times 10^7$ . (d) The mechanical mode with a quality factor of  $1.2 \times 10^3$  below the threshold of the mechanical instability. Inset shows the spectrum above the threshold of the mechanical instability with increasing pump power. Blue curves, experimental data; red curves, best fitting curves. (e) The spectrum of the Raman-laser mode in the 1676 nm band is read by the optical spectrum analyzer (OSA). The Raman-laser mode is below the threshold (blue curve) and excited (red curve) with increasing pump power. The inset shows the threshold curve of the Raman-laser mode.

mirror. The motion of the mirror changes the length of the cavity and thus induces an effective frequency shift to the optical cavity mode. The shift leads to effective optomechanical couplings with strength  $G_m$ . The coupling between the cavity modes and the molecular vibration mode can also be modeled as an optomechanical-type coupling, resulting in the formation of the Raman laser mode through their interaction. In such a model, the molecular mode interacts with the cavity modes just like a plasmonic cavity mode interacts with the optical mode.

As shown in Fig. 1(c), the quality factor of the optical mode is  $4.2 \times 10^7$ . The mechanical mode shown in Fig. 1(d) has a central frequency of 16.7 MHz and a mechanical quality factor of  $1.2 \times 10^3$  when the pump power is below the threshold of the mechanical instability, and the linewidth of the mechanical mode will be narrowed when the pump power is above the threshold. (see Fig. 1(d), inset). The frequency spectrum of the Raman-laser mode is shown in Fig. 1(e). The (blue) red curve represents the case that the Raman laser is (not) excited. Figure 1(e) shows the resonance peak of the Raman-laser mode at about 1676 nm. The inset of Fig. 1(e) displays the threshold curve of the Raman-laser mode.

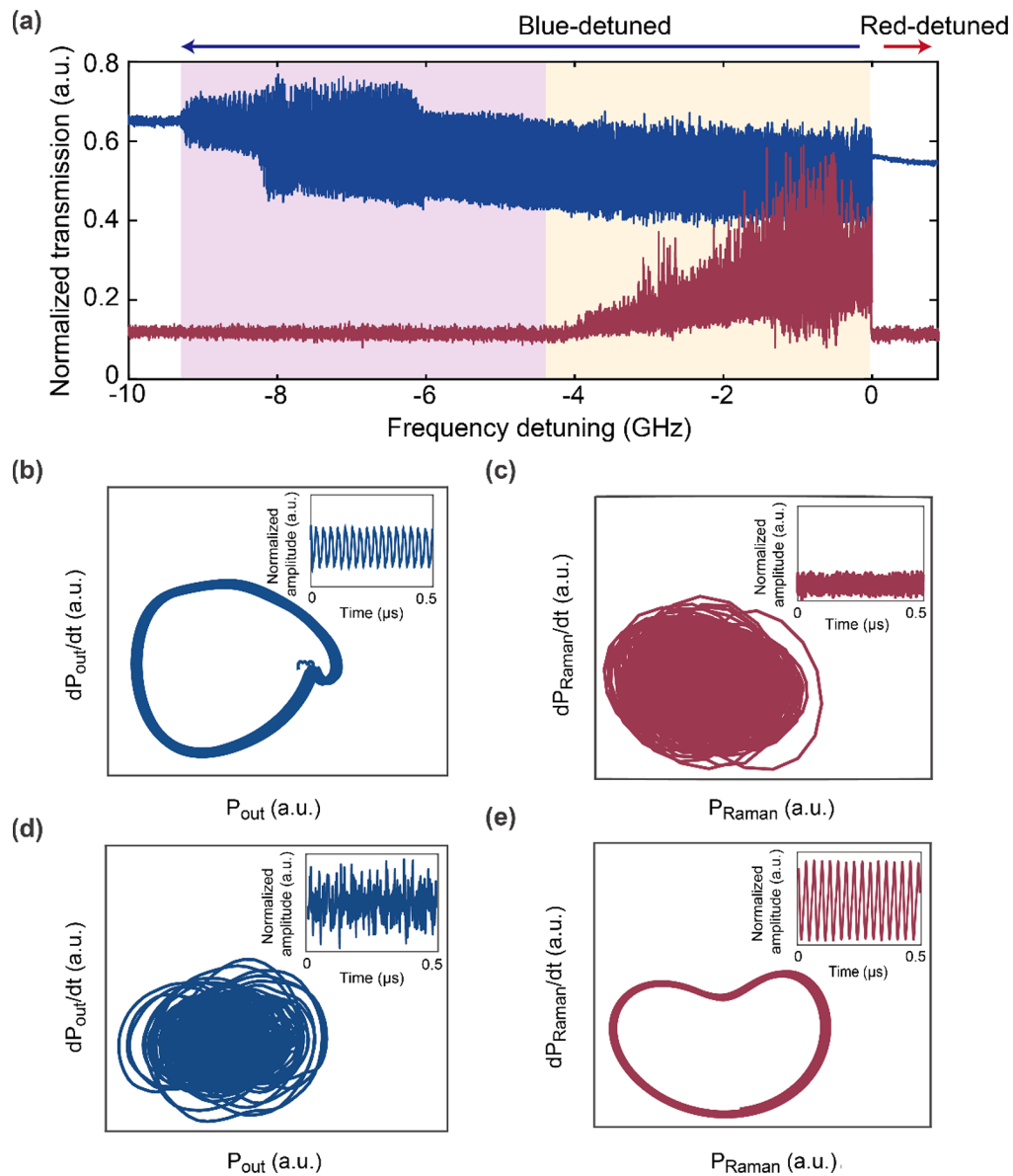
### 3. Order transfer from the optomechanical resonator to the Raman-laser mode

When varying particular system parameters, we can see the energy and order transfers between different components of the hybrid Raman-laser-optomechanical resonator. Figure 2(a) shows the time-domain signals of the optomechanical output field of the microtoroid resonator (upper blue curve) and the Raman-laser mode (lower red curve), respectively. By changing the frequency detuning between the input pump field and the cavity mode in the 1550 nm band, we obtain two different regimes in the blue-detuned regime. As the frequency detuning ranges from -4.3 GHz to -9.3 GHz (pink regime in Fig. 2(a)), the optomechanical motion of the resonator is periodic. Thus, this regime is referred to as the ordered mechanical regime. However, the Raman-laser mode is below the threshold, and the time-domain Raman-laser signal is disordered in this regime. When the frequency detuning exceeds -4.3 GHz (yellow regime in Fig. 2(a)), the phase transition takes place and new dissipative structures are gradually generated, under which the optomechanical mode is totally chaotic (disordered), and the Raman-laser mode is far above the threshold and thus generates an ordered output lasing field.

The phase transition process can be seen more clearly in the phase diagrams in Fig. 2(b-e), and the insets are the corresponding time-domain signals. In the ordered mechanical regime, when the frequency detuning is assigned as -6.8 GHz, as shown in Figs. 2(b, c), the phase diagram of the optomechanical mode is a single limit cycle and the Raman-laser mode is in a mess. This is confirmed by the sinusoidal time-domain signal of the optomechanical mode and the noisy time-domain signal for the Raman-laser mode, as shown in the insets of Fig. 2(b, c). When the frequency detuning is changed to -0.3 GHz, the system enters the ordered Raman-laser regime, while the optomechanical mode is totally chaotic (disordered), see Fig. 2(d). In contrast, the Raman-laser mode shown in Fig. 2(e) follows a single limit cycle and thus is periodic. In this regime, the order has been transferred from the optomechanical mode to the Raman-laser mode.

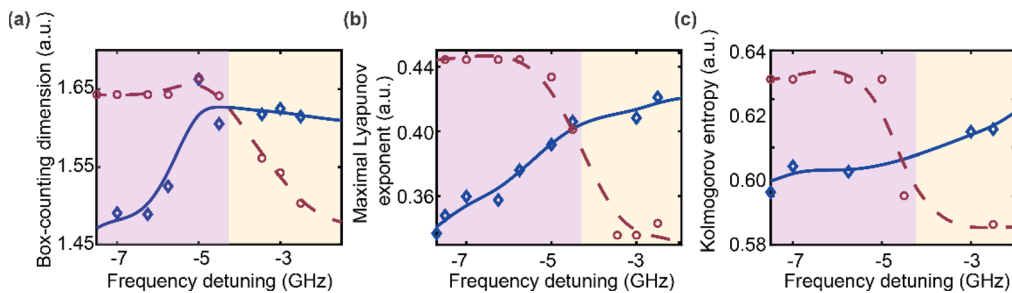
To give more insights for the above-illustrated phase transition process, we introduce different order parameters to quantitatively analyze the order transfer from the optomechanical resonator to the Raman-laser mode. Among all the order parameters, the box-counting dimension, the maximal Lyapunov exponent, and the Kolmogorov entropy are the most intuitive indicators in order transfer analysis. They also each have a rigorous criterion for determining whether a system is disordered. For example, integer and fractional values of the system's box-counting dimension correspond to ordered and disordered states, respectively. System order can also be determined by assessing whether the Kolmogorov entropy and the maximum Lyapunov exponent are greater than 0.

Let us first check the box-counting dimension of different system components, as shown in Fig. 3(a) (see the definition of the box-counting dimension in Supplement 1). The box-counting



**Fig. 2.** Time evolution of the hybrid Raman-laser-optomechanical resonator. (a) Different regimes are obtained by varying the frequency detuning between the pump mode and the cavity mode. The optomechanical mode (upper blue curve) and the Raman-laser mode (lower red curve) show two different regimes: the ordered mechanical regime (pink regime), and the ordered Raman laser regime (yellow regime). The phase diagrams are plotted by the derivatives of the output powers of the optomechanical mode and the Raman-laser mode versus the respective output powers. The phase diagrams of the optomechanical mode are presented in the ordered mechanical regime (b), the ordered Raman laser regime (d), and the corresponding phase diagrams of the Raman-laser mode are shown in (c) and (e).

dimension is a method used to quantify the spatial filling of a fractal set through the system phase diagram. Its fundamental concept involves evaluating how the count of boxes needed changes as the box size is adjusted. The box-counting dimension of an ordered structure is always an integer, while the disordered dissipative structure such as a chaotic attractor always has a fractal box-counting dimension. The box-counting dimension of the dissipative structure will increase when it turns to be more chaotic. In fact, we use the time series obtained in the experiment for calculation, so the time series are not perfectly periodic signals. The time series contains noise, so the box-counting dimension of the optomechanical mode is larger than 1 although the dynamics is a limit cycle. In Fig. 3(a), two different regimes are achieved by changing the frequency detuning. The box-counting dimension of the Raman-laser mode (red dashed curve) decreases and that of the optomechanical mode (blue solid curve) increases in the ordered mechanical regime. It means that order is gradually transferred from the optomechanical mode to the Raman-laser mode. When the frequency detuning is above the threshold of the Raman laser, that is in the ordered Raman laser regime, the box-counting dimension of the Raman-laser mode decreases rapidly and that of the optomechanical mode increases fast and stably. This means that the phase transition occurs and the periodic (ordered) limit cycle of the Raman-laser mode and the chaotic (disordered) attractor of the optomechanical mode are constructed. During this process, the chaotic optomechanical oscillation absorbs entropy from the Raman-laser mode by which the ordered Raman laser is generated.



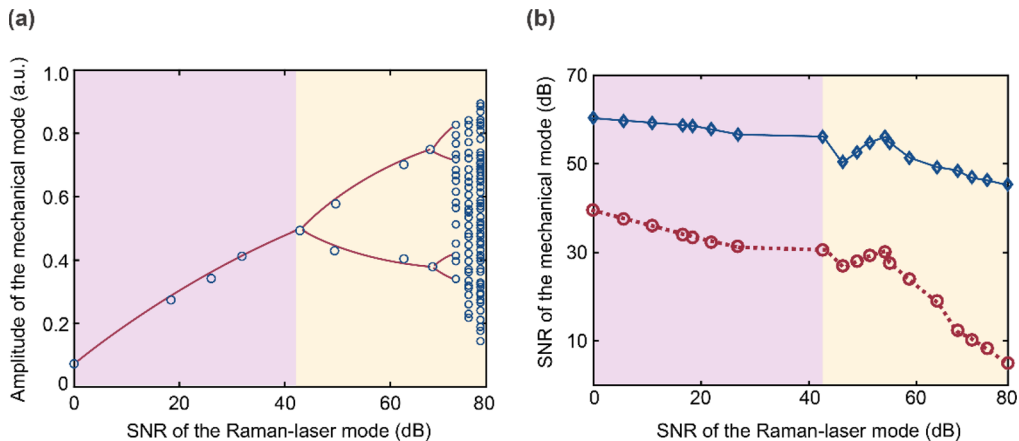
**Fig. 3.** Order transfer from the optomechanical mode to the Raman-laser mode is illustrated by different order parameters. (a) The box-counting dimension of the optomechanical mode (blue solid curve) and the Raman-laser mode (red dashed curve) versus the frequency detunings. (b) The maximal Lyapunov exponent of the optomechanical mode (blue solid curve) and the Raman-laser mode (red dashed curve) versus the frequency detunings. (c) The K entropies of the optomechanical mode (blue solid curve) and the Raman-laser mode (red dashed curve) versus the frequency detunings.

Another metric for characterizing the order of the system is the maximal Lyapunov exponent (MLE), which quantizes the derivation between different phase trajectories. The MLE is negative in an ordered system, while in a chaotic system, the MLE is positive. The more chaotic the system is, the larger MLE will become. The trajectories of MLE under different frequency detunings of the optomechanical mode (blue solid curve) and the Raman-laser mode (red dash curve) are shown in Fig. 3(b). Due to the measurement noise, the MLE is positive when the stationary trajectory of the system is a limit cycle. As the frequency detuning increases in the ordered mechanical regime, the optomechanical mode will gradually evolve from the periodic regime to the chaotic regime, while the signals of the Raman-laser mode become more orderly. Finally, the Raman-laser mode is sinusoidal and the hybrid system enters the ordered Raman-laser regime. In the ordered Raman-laser regime, the MLE of the optomechanical mode will remain higher than that of the Raman-laser mode.

The third parameter we introduce is the Kolmogorov entropy, which is also called K entropy in short. K entropy is a kind of entropy introduced to describe the degree of confusion (see the

definition of the K entropy in Supplement 1). It quantifies the rate at which trajectories diverge, indicating how quickly new information emerges as the system undergoes evolution. In the chaotic system, the K entropy is greater than 0. The larger the K entropy is, the more chaotic the system is. To characterize the two regimes, we tune the frequency detuning and check the K entropy of the hybrid Raman-laser-optomechanical resonator in Fig. 3(c). The time series contains noise, so the K entropy is larger than 0 although the system trajectory is a limit cycle. In the ordered mechanical regime, the K entropy of the optomechanical mode (blue solid curve) increases while that of the Raman-laser mode (red dashed curve) decreases, and the two curves intersect with each other on the boundary of the ordered mechanical regime. In the ordered Raman laser regime, the changes of the K entropy also show that a stable periodic limit cycle and chaotic attractor are constructed for the Raman-laser mode and the optomechanical mode respectively.

To better understand the formation process of the new dissipative structures, we further introduce the signal-to-noise ratio (SNR) to evaluate the degree of order of the system. In the experiment, whenever we change a frequency detuning, we can obtain the corresponding SNRs of the Raman-laser mode from the optical spectrum analyzer (OSA) and optomechanical mode from the electrical spectrum analyzer (ESA) respectively (see the experimental setup in Supplement 1). With the increase of the SNR, the system turns to be more ordered. In Fig. 4(a), in the ordered mechanical regime, with the increase of the SNR (order) of the Raman-laser mode, the amplitude of the optomechanical mode increases without bifurcation, which means that the phase transition does not occur in this case. In the ordered Raman laser regime, with the increase of the SNR of the Raman-laser mode, the optomechanical mode will begin to bifurcate, and finally, a stable chaotic attractor is generated.



**Fig. 4.** Order transfer between the optomechanical mode and the Raman-laser mode in two different regimes. (a) The bifurcation diagram of the optomechanical mode versus the SNR (order) of the Raman-laser mode. Period-doubling bifurcation occurs for the optomechanical mode with the increase of the SNR of the Raman-laser mode. (b) The SNR of the optomechanical mode at the main peak (blue solid curve) and the period-doubling peak (red dashed curve) versus the SNR of the Raman-laser mode in the ordered mechanical regime, and the ordered Raman laser regime respectively.

In Fig. 4(b), the SNR of the optomechanical mode at the main peak (blue solid curve) and the period-doubling peak (red dashed curve) changes with the increase of the SNR of the Raman-laser mode. In the ordered mechanical regime, the SNR of the optomechanical mode decreases gradually with the increase of the SNR of the Raman-laser mode, which means that order is gradually transferred before the phase transition occurs. In the ordered Raman laser regime, the

SNR of the optomechanical mode stably decreases further with the increase of the SNR of the Raman-laser mode, which means that the order is transferred from the Raman-laser mode to the optomechanical mode further and new dissipative structures are generated. Additionally, we can see from Fig. 4(b) that the SNR of the optomechanical mode at the period-doubling peak (red dashed curve) changes more drastically than that at the main peak (blue solid curve), which means that the SNR of the optomechanical mode at the period-doubling peak is more sensitive and thus a more powerful tool to describe the order transfer between the two modes in the two different regimes.

#### 4. Conclusion

In conclusion, we experimentally demonstrated that an ordered Raman laser can be generated via the entropy absorption of a disordered chaotic source. We introduced the box-counting dimension, the Kolmogorov entropy, and the SNR of different components, to quantitatively analyze the order transfer from the optomechanical mode to the Raman-laser mode in the hybrid Raman-laser-optomechanical resonator. It was found that, in this ordered optomechanical system, the phase transition occurred and new dissipative structures were constructed during the order transfer process. The period-doubling bifurcation process of the optomechanical mode was experimentally observed. The steeper curve of the SNR of the period-doubling peak means better sensitivity for evaluating order transfer. Our work showed a new mechanism for realizing and controlling on-chip micro-lasers.

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**Disclosures.** The authors declare no conflicts of interest.

**Data availability.** Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

**Supplemental document.** See [Supplement 1](#) for supporting content.

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