Raman-lasing dynamics in split-mode microresonators

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(Received 18 November 2013; published 5 January 2015)

We study the dynamics of stimulated Raman scattering in high-$Q$ split-mode microresonators. It is found that Raman lasing preferentially occurs in only one of the two competing split modes, and can therefore be beat-note free. Sweeping the pump wavelength causes the intracavity field pattern of the pump light to rotate, which makes the Raman laser hop between the two split modes. During these hops, both modes lase simultaneously, producing a beat note in the output Raman power. By studying this beat note, we demonstrate that the mode splitting is strongly affected by the Raman intensities in the two modes via the optical Kerr effect.

DOI: 10.1103/PhysRevA.91.013804 PACS number(s): 42.55.Ye, 42.79.Gn

I. EXPERIMENTAL RESULTS

Figure 1(a) shows the schematic of the experimental setup in which a fiber taper [30] is used to couple the pump light (680-nm wavelength band; power $\sim$1.1 mW) into a microsphere and to collect the emission. The microspheres were built on a silicon wafer [31,32] and have radii $R \sim 15 \mu$m, allowing for small mode volumes of $\sim$100 $\mu$m$^3$ and high $Q$ factors exceeding 10$^7$. We use microspheres instead of microtoroids to suppress optomechanical oscillations [33]. When the pump laser is scanned through a cavity resonance (pump mode), the transmission spectrum [Fig. 1(b)], the first-order Raman spectrum [Fig. 1(c)], and the Raman emission in the time domain [Fig. 1(d)] are recorded. The transmission spectrum exhibits a much larger broadening than its original linewidth during the pump wavelength up-scan (speed: 5.4 nm/s or $\sim$3.5 THz/s), due to thermal effects. The transmission spectrum at low power is shown in the inset, from which we see a doublet with an intrinsic splitting of $2g/2\pi \sim 75$ MHz and $Q$ factor of $\sim 4.6 \times 10^7$.

Unlike conventional rare-earth-ion doped microcavity lasers in which a nearly constant beat note in the output laser power is expected during the entire pump scan, here we observe periodic pairs of short occurrences $A$ and $B$ (durations $\sim 5 \mu$s; period $\sim 67 \mu$s) of a beat note (average frequencies of $\sim$55 MHz), separated by a linear increase of Raman power with no beat note, as shown in Fig. 1(d). A closer view reveals that the beat frequency either increases or decreases monotonously in each occurrence, and that the sign of the frequency change alternates from one occurrence to the next. For instance, if the beat note speeds up during occurrence $A$, it then slows down in the next occurrence $B$.

To confirm that the observed Raman-lasing behavior in Fig. 1(d) is common in split-mode microcavities, we study the Raman emission in the time domain under different experimental conditions. First we gradually increase the pump power, and measure the Raman emission from the microcavity, with the result displayed in Fig. 2, in which the input power is gradually increased from Fig. 2(a) to 2(d). It can be seen that for different pump powers, as long as the power is large enough to enable stimulated Raman emission, the temporal behavior is similar; the emission profile exhibits the same

Stimulated Raman scattering (SRS) [1] holds great promise for extending the wavelength of existing lasers [2], generating ultra-short light pulses [3], and developing new biomedical imaging techniques capable of chemically specific diagnostics of molecular species [4,5]. Over the past decade, high-$Q$ optical microcavities have been proved to be an ideal tool for enhancing SRS [6–17]. For instance, whispering-gallery-mode (WGM) microcavity Raman lasers with thresholds as low as 3 $\mu$W and photon conversion efficiencies exceeding 60% have been realized [18,19]. In such microcavities, the two initially degenerate counterpropagating traveling-wave modes inevitably couple to each other and produce two new split modes [20–23], strongly affecting the intracavity optical behavior. In the case of microcavities doped with active materials such as rare-earth ions, these split modes are responsible for the emergence of a beat note in the lasing emission [24,25], which, despite applications for nanoparticle detection [26–29], can also be a limitation of such cavities.

In this paper, we study the dynamics of Raman lasing in the split-mode regime, and show several interesting phenomena. We first present our experimental results, and propose that, independently of whether Raman lasing occurs, scanning the frequency of the pump laser across a cavity resonance causes the field pattern of the pump light inside the cavity to rotate. We then demonstrate that strong mode competition in Raman lasing generally leads to lasing in only one split mode, as opposed to rare-earth-ion doped microcavity lasing where both modes lase. Resulting from the rotation of the field pattern of the pump light, laser hopping occurs between the two split Raman modes, and the active mode(s) can be selected. Furthermore, during each hop, both split modes lase simultaneously, and we show by monitoring the beat note that the mode splitting changes significantly during the hop. We numerically demonstrate that this phenomenon is caused by the different light intensities in the two Raman modes via the optical Kerr effect.

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1050-2947/2015/91(1)/013804(6) 013804-1 ©2015 American Physical Society
FIG. 1. (Color online) (a) Schematic of the taper-microsphere coupling system. The output light is divided into three parts to measure (b) the transmission spectrum, with the inset showing the case at low pump power (frequency detuning and time are both shown, since they are proportional during the pump frequency scan), (c) the Raman spectrum, and (d) the temporal Raman power, with the inset showing several typical occurrences of the beat note.

FIG. 3. Raman emission in the time domain using a pump laser in the 1550-nm wavelength band. Inset: The corresponding pump transmission spectrum, which exhibits oscillations.

periodic occurrences of a beat note. Increasing the taper-cavity coupling strengths from under coupling to critical coupling, also gives similar results, as shown in in Figs. 2(e)–2(h). Note that the observed beat note is different from the optical ringing effect [34–37], which can also be observed in such cavities [inset of Fig. 2(a)], and that the appearance of second- and third-order Raman lasing changes the slope of the emission. Similar results are consistently obtained for different-sized microcavities.

We also study the temporal Raman-lasing behavior using a different pump laser, in the 1550-nm band. In the measurement, a 1550/1670-nm wavelength division multiplexer (WDM) is used to separate the pump and Raman lasers. The Raman emission in the time domain is shown in Fig. 3, and the
corresponding transmission spectrum is plotted in the inset. We observe the same phenomena as with a 680-nm pump: periodic occurrences of a beat note in an otherwise monomode emission profile. These results confirm that the behavior consistently occurs in split-mode microcavities.

Note that the transmission spectrum also exhibits periodic oscillations, with the same oscillation period as the Raman mode hopping, as shown in the inset. These oscillations start before the onset of Raman lasing and continue throughout the frequency scan. Similar results have also been obtained in the 680-nm band, as shown in Fig. 1(b). The physical origin of these oscillations in the transmission spectrum will be further discussed in the next section.

II. ROTATION OF THE PUMP FIELD PATTERN

To explain our experimental results, we first propose that when the frequency of the pump laser is scanned across a cavity resonance in a split-mode microcavity, the field pattern of the pump light in the cavity rotates. When the pump light is coupled into the cavity in the clockwise (CW) direction, the CW wave builds up first, followed by the counterclockwise (CCW) wave due to backscattering. There is therefore a time delay \( t \) between the buildups of the CW and CCW waves. This time delay is inversely proportional to the coupling strength between the two waves. Due to the scanning action of the pump laser, this time difference \( t \) corresponds to a frequency difference \( \Delta \Omega \) between the CW and CCW waves [Fig. 4(a)], which is roughly \( t \) times the scanning speed of the pump laser. As is the case in ring laser gyroscopes [38], the frequency difference between the two counterpropagating waves causes the interference pattern to rotate inside the cavity, with a period \( T = 1/\Delta \Omega \) between two identical configurations of the intracavity light intensity.

This rotation of the pump mode inside the cavity causes the overlap between the intracavity field and the fiber taper used for coupling to change over time, which leads to the observed oscillations in the transmission spectrum of the pump laser, as well as in the Raman power at the output. Note that at low pump powers, at which the mode linewidth is not thermally broadened, these oscillations in the transmission cannot be observed, since the oscillation period (67 \( \mu \)s with our experimental parameters) is much longer than the time during which the pump light would then be coupled into the cavity.

To confirm that the observed oscillations are indeed caused by a rotation of the field pattern of the pump laser, we first changed the scanning speed of the pump frequency. As we expected, the frequency of the oscillations, and of the occurrences of a beat note, was proportional to the scanning speed [Fig. 4(b)]. Furthermore, when we assume \( r = 1/g \) with \( g \) being the backscattering strength in a single-scatterer model, and use the experimental parameters \( v = 3.5 \) THz/s for the pump scanning speed and \( g/2 \pi = 37.5 \) MHz corresponding to the observed mode splitting for a given cavity, the frequency difference \( \Delta \Omega \) is estimated to be \( \sim 15 \) kHz, which agrees well with the experimentally observed oscillation frequency of 15 kHz.

III. MODE COMPETITION AND SELECTIVE SPATIAL MODE EXCITATION

In our experiments described in the first part, the beat note of the Raman laser is not present most of the time, unlike doped microcavity lasers where both modes are usually active. We attribute this difference to the distinct mode competition mechanisms. Since the random surface roughness induced split pump mode is not perfectly sinusoidally distributed along the azimuthal direction [39], the spatial overlaps between the pump mode and the two split lasing modes are not exactly equal, leading to slightly different thresholds.

In microcavities doped with active materials such as rare-earth ions, since the two nearly orthogonal split lasing modes have quite different field distributions, dopants located at different sites can therefore contribute differently to the two lasing modes. The two split modes obtain gain from dopants located in their own mode volumes, so gain clamping only occurs once both modes are lasing. Mode competition in this case is weak, and both split modes can and usually do lase simultaneously even with different thresholds for the two split modes.

In Raman lasing, however, it is the intracavity pump intensity that is clamped at its threshold value. As opposed to doped microcavity lasing where different ions contribute to lasing in the two modes, both Raman modes share exactly the same pump photons. When the lower-threshold Raman mode lases, the number of pump photons in the cavity is clamped, effectively clamping the gain for both modes. As the pump power increases, any additional input photons are converted to Raman photons in the low-threshold mode. The lasing threshold in the other split Raman mode is therefore not reached, and only one mode can lase. This is indeed what we observe most of the time in our experiment.

The unexpected part of the temporal Raman emission described in the first part is the periodic occurrence of a beat note. The frequency at which this beat note appears is equal to that of the oscillations in the transmission caused by the rotation of the pump mode. We propose that this periodic

![FIG. 4. (Color online) (a) Theoretical spectra of the CW and CCW waves in the resonator under our experimental conditions (pump scanning speed of 3.5 THz/s), relative to the instantaneous central frequency of the pump laser during the scan. (b) The dependence of the Raman-lasing hopping frequency on the pump scanning speed. (c) and (d) Raman intensities when the pump light is locked at two different wavelengths.](013804-3)
occurrence is a consequence of the Raman laser periodically hopping between the two split modes, due to the rotation of the pump field pattern.

The Raman modes are not subject to the same rotation as the pump mode, since SRS occurs simultaneously in both directions. As a result, the spatial overlaps between the rotating pump mode and the two nonrotating Raman modes switch periodically. Since the difference in the thresholds of the two Raman modes is mostly caused by the difference in overlap between the Raman and pump modes, the rotation of the pump mode causes the Raman thresholds to alternate, causing the observed mode hopping. During each hop, both Raman modes lase simultaneously, thus producing a beat note. As the hopping occurs twice during the time necessary for the pump field to return to its original distribution, two hops are observed during every period of the oscillations in the transmission.

Experimentally, this behavior can be controlled; locking the wavelength produces either a stable-frequency beat or a stable monomode Raman emission without oscillations, as shown in Figs. 4(c) and 4(d), respectively. If the lock occurred during a cross-over when both modes have the same threshold, both modes can be made to lase, and a constant beat note appears. Otherwise, only one mode lases.

IV. KERR EFFECT INDUCED CHANGE IN THE MODE SPLITTING

We show here that the optical Kerr effect is what causes the change of beat frequency during the hops, and that the mode splitting is strongly affected by the distribution of Raman intensities between the two modes. When the Raman lasing hops from the shorter-wavelength mode \( R_− \) to the other \( R_+ \), due to the Kerr effect, \( R_− \) is blue-shifted while \( R_+ \) is red-shifted, so the mode splitting increases and the observed beat oscillation therefore accelerates. Conversely, when lasing hops from \( R_+ \) to \( R_− \), the beat frequency slows down. This is exactly what we observe in Fig. 1(d). Confirming this process, Fig. 5(d) shows the beat frequency of the Raman power by performing a local fast Fourier transformation (FFT) to Fig. 1(d). The amplitude of the beat frequency change scales linearly with the Raman power, because of the linear dependence of the refractive index change on the Raman power.

We now quantitatively study the Raman-lasing dynamics to demonstrate that the Kerr effect is indeed responsible for the change in the mode splitting. We first obtain the evolution of the intracavity pump and Raman fields, then calculate the Raman mode splitting by taking the Kerr effect into account, and finally derive the Raman emission from the microcavity. The rate equations for pump and Raman modes [10] are

\[
\frac{da_P}{dt} = -\left( \frac{1}{2\tau_P} + \sum_{j=\pm} \omega_P G_{R_j} |a_{R_j}|^2 + i\Delta \omega \right) a_P + \frac{\kappa a_{in}}{\sqrt{2}},
\]

\[
\frac{da_{R_j}}{dt} = -\left( \frac{1}{2\tau_{R_j}} - G_{R_j}^c |a_P|^2 \right) a_{R_j},
\]

where \( a_P \) and \( a_{R_j} \) represent the amplitudes of the pump and Raman modes, respectively, with resonance frequencies \( \omega_P \) and \( \omega_{R_j} \), and photon lifetimes \( \tau_P \) and \( \tau_{R_j} \). \( a_{in} \) denotes the input light with a coupling strength \( \kappa \) and a detuning \( \Delta \omega \) with the pump mode. \( G_{R_j}^c = \frac{f_{R_j}^c c}{2\pi V_P} \) is the Raman gain for \( R_{\pm} \), where \( f_{\pm}, c, G_R, n, \) and \( V_P \) denote the spatial overlap between the pump mode and the Raman mode \( R_{\pm} \), the speed of light in vacuum, the bulk Raman gain of the cavity material, the refractive index, and the pump mode volume. The difference of the overlaps \( (f_+ - f_-) \) between the pump mode and the two split Raman modes, which represents the degree of deviation of the pump mode distribution from a perfect sinusoidal wave, cannot be directly obtained from experiment or theory. To simulate the periodic threshold fluctuations for the two lasing modes caused by the alternating overlaps, we therefore add a time-dependent sinusoidal component with a 1% amplitude (after fitting) and a period of 67 \( \mu \)s (corresponding to that of both the experimentally observed and theoretically calculated hopping period) to the relative gain \( G_{R_j}^c \). The detuning \( \Delta \omega \) is

![FIG. 5. (Color online) (a) Simulation results for the intracavity energies in the pump (black solid curve) and Raman (red dashed and blue dotted curves) modes. (b) and (c): The corresponding beat frequency and Raman emission from the microcavity. Insets of (a) and (c): the corresponding closeups, with four different evolution ranges. (d) Local FFT of the beat frequency change in each occurrence of the experimental Raman power in Fig. 1(d).](image-url)
obtained through the thermal evolution of the pump mode

\[
\frac{d\Delta T}{dt} = -\gamma_{th}\Delta T + \left( \frac{Q}{Q_{abs} a_P} \right) |a_P|^2 \\
+ \sum_{j=+,-} 2 \left( \frac{\alpha_{jP}}{\alpha_{jR}} - 1 \right) G_{jR}^n |a_{jR}|^2 |a_P|^2,
\]

where \(\Delta T\) and \(C\) are the temperature change and thermal capacity of the mode volume, and \(\gamma_{th}\) represents the thermal conductivity between the mode volume and environment. \(Q\) and \(Q_{abs}\) are the total and material-absorption-limited \(Q\) factors, with \(Q/Q_{abs}\) thus being the proportion of intracavity pump photons that are converted to heat. This equation includes the contribution to heating from both the silica absorption [40] and the inelastic Raman scattering [41], and we find that the latter is dominant. The variation in the Raman mode splitting due to the Kerr effect is given by \(\Delta f = c^2 n_2 (|a_{R+}|^2 - |a_{R-}|^2)/(\lambda R n_2 V_{mode})\), where \(n_2 = 1.46\) and \(n_2 = 2.7 \times 10^{-16} \text{cm}^2/\text{W}\) are the linear and nonlinear refractive indices of silica at the Raman wavelength \(\lambda_R = 707\) nm, \(V_{mode}\) is the Raman mode volume, and \(c\) is the speed of light in vacuum. In the simulation, we use the experimental parameters in Fig. 1. A thermal loss rate \(\gamma_{th}/C = 7 \times 10^4 \text{s}^{-1}\) yields the experimental Raman emission profile.

The intracavity energies in the pump and Raman modes are plotted in the solid, dashed and dotted curves in Fig. 5(a). The evolution of the mode splitting and the corresponding Raman emission are plotted in Figs. 5(b) and 5(c). These results are consistent with the experimental observations. Single-mode lasing occurs when the threshold of one mode is above the other [regions I, III, and V in the insets of Figs. 5(a) and 5(c)], and the hopping occurs when their thresholds cross over one another (regions II and IV). For example, region II corresponds to a hop from \(R_+\) to \(R_-\), and region IV to a hop from \(R_-\) to \(R_+\). The mode splitting given by the simulation agrees well with the experimental beat frequency in Fig. 5(d). Note that the influence of the Kerr effect is particularly strong. For instance, with the experimental Raman power of \(\sim 60\) \(\mu\)W, the mode splitting changes from \(\sim 27\) MHz to \(\sim 82\) MHz. Also note that the thermal nonlinearity also affects the mode splitting; however, due to the fast thermal exchange rate between the two modes, we found its contribution to be negligible compared to the Kerr effect.

V. CONCLUSION

To conclude, we have studied the dynamics of Raman lasing in split-mode microcavities and have shown that several interesting phenomena occur. First of all, independent of Raman lasing, scanning the frequency of the pump laser causes the field pattern of the pump light to rotate inside the microcavity. Raman lasing generally occurs in only one mode due to strong mode competition, as opposed to lasing in microcavities doped with active materials such as rare-earth ions. Furthermore, results from the rotation of the field pattern of the pump mode inside the cavity, the active Raman lasing mode(s) can be selected. Finally, we showed that the magnitude of the mode splitting was strongly affected by the optical Kerr effect, due to the unequal intensities of Raman light in the two split modes. We believe that our work contributes to better understanding and controlling the properties of Raman lasing in ultrahigh-\(Q\) microcavities.

ACKNOWLEDGMENTS

We thank Xiao-Chong Yu for the help with the data processing. William R. Clements and Bei-Bei Li contributed equally to this work. This work was supported by the NSFC (No. 61435001, No. 11474011, No. 11222440, and No. 1121091), the 973 program (No. 2013CB328704), and the Beijing Natural Science Foundation Program (No. 4132058). B.Q.S. was supported by the National Fund for Fostering Talents of Basic Science (Grants No. J1030310 and No. J1103205).