MHz-level self-sustained pulsation in polymer microspheres on a chip
Zhou-Chen Luo, Cao-Yuan Ma, Bei-Bei Li, and Yun-Feng Xiao

Citation: AIP Advances 4, 122902 (2014); doi: 10.1063/1.4903317
View online: http://dx.doi.org/10.1063/1.4903317
View Table of Contents: http://scitation.aip.org/content/aip/journal/adva/4/12?ver=pdfcov
Published by the AIP Publishing

Articles you may be interested in
Anisotropic wrinkle formation on shape memory polymer substrates

Thermochromic polymer opals
Appl. Phys. Lett. 95, 173116 (2009); 10.1063/1.3256193

Effect of polymer concentration on stabilized large-tilt-angle flexoelectro-optic switching

Conditions for self-sustained pulsation and bistability in semiconductor lasers
J. Appl. Phys. 58, 1689 (1985); 10.1063/1.336065

Self-sustained pulsations in semiconductor lasers: experimental results and theoretical confirmation
J. Appl. Phys. 51, 4029 (1980); 10.1063/1.328227
MHz-level self-sustained pulsation in polymer microspheres on a chip

Zhou-Chen Luo,1 Cao-Yuan Ma,1 Bei-Bei Li,2,a and Yun-Feng Xiao1,3,b
1State Key Laboratory for Mesoscopic Physics & School of Physics, Peking University, Beijing 100871, P. R. China
2Qian Xuesen Laboratory of Space Technology, Beijing 100094, P. R. China
3Collaborative Innovation Center of Quantum Matter, Beijing 100871, P. R. China

(Received 30 September 2014; accepted 22 November 2014; published online 2 December 2014)

We observe MHz-level periodic self-sustained pulsation (SSP) in the transmission spectrum of a polydimethylsiloxane (PDMS) spherical microcavity on a silicon chip, under a fixed-frequency continuous laser excitation. The SSP results from the strong competition between the thermo-optic and thermal expansion effects of PDMS within the cavity mode volume. The experimental results show good agreement with the theoretical prediction by considering the modification of the thermal expansion coefficient and the temperature distribution within the mode volume.

I. INTRODUCTION

Whispering gallery mode (WGM) microresonators featured by ultra-high Q factors and small mode volumes are promising for a variety of applications, such as for sensing,2–9 microlasing,10–12 cavity quantum electro-dynamics,13–15 and for cooling of mechanical systems.16–19 Studies have shown abundant nonlinear behaviors promoted by the highly localized optical field inside the microcavities, such as optical bi-stability,20,21 deformation and abnormal oscillation22–27 of the transmission spectra. Recently, periodic self-sustained pulsation (SSP)28–32 in the transmission spectrum has been reported in various microcavity systems, excited by a fixed-frequency continuous laser, different from that excited by frequency-scanned lasers26 or by pulsed lasers27 in previous studies. For instance, very fast SSPs at the GHz level have been reported in silicon microcavities28,29 resulting from coupled electron-photon dynamics. Moreover, in silica microcavities or silica/polymer hybrid microcavities where there are no free carriers, SSPs have also been observed, formed by the interplay between thermo-optic effect and Kerr effect,30 or between thermo-optic effect and mechanical motion.31,32 The phenomenon of SSP in various WGM microresonators provides a simple optical oscillator, which for example can be used to generate continuous pulse lasers in Er-doped microcavities33 and has also led to certain other sensing applications.32 Generally, the SSPs in silica or silica/polymer microcavities have relatively small frequencies, usually at the Hz or kHz level.30–32 In this work, we report SSP in a polymer microsphere on a silicon chip, excited by a fixed-frequency continuous laser, with a MHz-level oscillation frequency, much faster than previously reported silica or silica/polymer microcavities. A theoretical model is built to interpret the physical origin of the SSP, based on the thermal and optical dynamics of the cavity, by taking into account the modification of the thermal expansion coefficient and the temperature distribution within the mode volume. It is revealed that the fast SSP in the polymer microcavity is induced by the periodic oscillation of the cavity resonance frequency via the competing thermo-optic and thermal-expansion effects of the polymer within the mode volume. The effects of the input power
and the frequency of the excitation laser on the pulse period are further studied to validate our theoretical model.

II. EXPERIMENTAL SETTINGS AND OBSERVATIONS

To obtain the on-chip polydimethylsiloxane (PDMS) microsphere, we first fabricate a silica microsphere on a chip.\textsuperscript{34,35} We then use the method introduced in previous works\textsuperscript{26,35,36} to coat a layer of PDMS onto the surface of the silica microsphere. Compared with the spin-coating method,\textsuperscript{37} this droplet-coating method enables coating a specific microcavity with a certain thickness. Figure 1(a) gives the top-view optical image of the microsphere before (left half) and after (right half) coating, from which we can see that the silica microsphere has a diameter of $\sim 27$ $\mu$m, with a coating layer of $\sim 2.3$ $\mu$m. With such a thick coating layer, almost all the field energy distributes into the PDMS layer, as shown in the normalized field distribution of the fundamental mode in the cross section of the microcavity in Fig. 1(c). Since all the nonlinear effects (including the optical Kerr effect, thermo-optic and thermal expansion effects) are proportional to the local optical power, the temperature variation in silica can be neglected compared with that in the PDMS layer, and the nonlinear effects of the silica core have little contribution to the dynamic behavior of the hybrid microsphere. Therefore, this hybrid microcavity can be regarded as a pure PDMS cavity, different from the cases in previous works\textsuperscript{26,32} in which the thermal effects of both the silica core and the polymer coating layer should be considered.

To test the thermal dynamics of the PDMS microcavity, we use a tunable laser in the 680 nm wavelength band as the probe laser. A fiber taper with a diameter of $\sim 500$ nm is used to efficiently couple the laser light into the cavity and also to collect the output, as displayed in the side view optical image of the coupling system in Fig. 1(b). A triangular wave voltage is exerted to the piezoelectric transducer (PZT) of the tunable laser to tune the wavelength. Due to the high Q factor ($\sim 1 \times 10^7$) and the large thermal effect of PDMS, the laser light can be coupled into the microcavity for a large range of wavelength. We then gradually decrease the amplitude of the triangular wave voltage to zero (i.e., stop scanning the laser wavelength), and record the transmission in the time domain. The taper-cavity gap and the input polarization are precisely adjusted to obtain a critical coupling between the fiber taper and the microcavity. Under an input power of $\sim 1.2$ mW, periodic pulses emerge in the transmission, as shown in Fig. 2(a), with its close-up in the marked region plotted in Fig. 2(b). The pulsation can be consistently maintained under a stable experimental environment. We also study the behavior of the microcavity using a different pump laser, in the 780 nm wavelength band, and observe similar result. The period of the pulsation is of the order of microsecond, much smaller than that in previous reports of mechanical movement involved SSP on silicon nitride microdisks (of the order of second) (Ref. 31), and PMMA-coated silica microtoroids (of the order of millisecond) (Ref. 32). This difference will be analyzed in the following part.
III. THEORETICAL MODEL

We now turn to study the physical origin of the fast SSP in the PDMS microcavity. The phenomenon of SSP or similar optical bi-stability in a variety of optical devices was demonstrated to be induced by two competing nonlinear effects with comparable magnitude, but different timescales.\textsuperscript{28–32} To simplify and generalize the theoretical model, previous works generally characterized the whole cavity mode volume with one (for a cavity with only one material) or two (for a cavity composed of two materials) phenomenological temperatures and neglect the temperature distribution within the cavity mode volume. However, for the PDMS microcavity, the temperature distribution inside the mode volume is the key to the formation of SSP, which is briefly explained as follows. When the temperature of the PDMS cavity increases, the negative thermo-optic effect (thermo-optic coefficient $n_1 = dn_1/dT = -1 \times 10^{-4} \text{ K}^{-1}$) and the positive thermal expansion effect (thermal expansion coefficient $n_2 = (1/R)dR/dT = 2.7 \times 10^{-4} \text{ K}^{-1}$) shift the cavity resonance to opposite directions. For the thermo-optic effect, the inner layer where the field intensity is strongest has the largest contribution to the blue shift of the cavity mode resonance. For the thermal expansion effect, however, due to the non-uniform increase of the temperature within the cavity mode volume, the expansion of the hotter region (inner layer) is inevitably restricted by the colder region (outer layers). The real thermal expansion coefficient of PDMS should be modified into $\alpha(\vec{r}) \times n_2$, where $0 < \alpha(\vec{r}) < 1$ is the modification factor with a smaller value for the inner layer. Therefore, the thermo-optic effect blue shifts the cavity resonance first when the light is coupled into the cavity, and then the thermal expansion effect pulls the resonance back later after the heat is conducted to the outer layers where $\alpha(\vec{r})$ are larger. This induces a periodic oscillation of the cavity mode resonance, leading to the SSP in the transmission. The resonance shift of the cavity mode induced by the temperature variation is
\[ \delta \omega(t) = -\omega_0 \times \iiint_V \left[ (n_1 + \alpha(\tilde{r}) \times n_2) \times \Delta T(\tilde{r}, t) \times \eta(\tilde{r}) \right] dV, \] (1)

where \( \eta(\tilde{r}) = \frac{n(\tilde{r})^2 |E(\tilde{r})|^2}{\iiint_{V} |n(\tilde{r})^2 |E(\tilde{r})|^2|dV} \) is the field energy density at the position \( \tilde{r} \). Since the temperature within the mode volume is much higher than that of the rest part of the cavity, we only consider the temperature change within the mode volume. To simplify the numerical calculation of the integral in Eq. (1) and meanwhile achieve agreeable precision, we divide the light spot region (where it has field distribution in Fig. 1(c)) into 11 layers according to the contour of the optical field density, labeled with \( i = 0, 1, \cdots, 10 \), from the innermost to the outermost of the light spot. The relative optical density in the 0th layer is 28.6 and 2.6 in the 10th layer. The region with relative optical density lower than 2.6 has no significant temperature variation, and is omitted. The temperature variation of each layer is denoted by \( \Delta T_i(t) \) and the modification factor \( \alpha_i \) is assumed to be uniform inside one single layer and is used as a fitting parameter here. Then Eq. (1) can be simplified into

\[ \Delta \omega(t) = -\omega_0 \times \sum_{i=0}^{11} (n_1 + \alpha_i \times n_2) \times \Delta T_i(t) \times \eta_i, \] (2)

where \( \eta_i \) is the field energy proportion in the \( i \)th layer and can be calculated theoretically. The overall shift of the cavity resonance frequency is the weighted sum induced by the 11 PDMS layers. The governing equations of the dynamical behavior of the temperature variation and optical field are

\[ \frac{d \Delta T_i}{dt} = \frac{1}{C_T \rho S_i L} \left[ -K_h \times S_i \times \nabla T_i + \alpha_{\text{abs}} \times P_C \times \eta_i \times L \right], \] (3)

\[ \nabla T_i = \left( \frac{\Delta T_i - \Delta T_{i-1}}{d_{i-1}} + \frac{\Delta T_i - \Delta T_{i+1}}{d_i} \right), \] (4)

\[ \frac{dE_C(t)}{dt} = -[\delta_0 + \delta_C + i\Delta \omega(t)] E_C(t) + i\frac{K}{\tau_C} E_{in}. \] (5)

Equation (3) and (4) is the simplified Fourier law of heat conduction. All the constants can be found in Refs. 26, 36, 38, and 39: \( C_p = 1300 \text{ J/kg/K} \) and \( \rho = 965 \text{ kg/m}^3 \) are the heat capacity and mass density of PDMS, respectively. \( K_h = 0.2 \text{ W/m/K} \) is the heat conductivity coefficient, \( \alpha_{\text{abs}} = 3.45 / m \) is the linear absorption coefficient, \( S_i \) is the cross sectional area of the \( i \)th layer, \( L = \pi D \) is the perimeter of the whole microsphere, \( d_i \) is the distance between the center of two adjacent layers. \( P_C(\tau) = |E_C(\tau)|^2 / \tau \) is the circulating power inside the cavity, with \( \tau = \frac{\eta_0 D}{c} \sim n_0 \pi D/c \) and \( \omega_0 = \omega_0 / 2Q_0 \) and \( \delta_C = \omega_0 / 2Q_C \) are the intrinsic loss and the coupling induced loss of the cavity mode, respectively, with \( Q_0 \) and \( Q_C \) denoting the intrinsic and coupling Q factors, respectively. \( \kappa = \sqrt{2\delta_C \tau_C} \) is the coupling coefficient, \( |E_{in}| = \sqrt{P_{in} / \tau_C} \) is the input optical amplitude, with \( P_{in} \) being the input laser power. The temporal behaviors of the optical field and temperature variation of the PDMS cavity are determined by the above three equations and can be solved through iteration method.

Figure 3(a) displays the calculated transmission of the PDMS microcavity, which shows a good agreement with the experimental result in Fig. 2(b). In Fig. 3(b) we plot the temperature change in each layer, shown in the 11 colored curves. In order to analyze the process, we divide one SSP period into four stages, marked by four different colors in Figs. 3(a) and 3(b). (i) When the cavity resonance is slightly below the frequency of the input laser, the optical energy begins to be coupled into the cavity and the cavity is heated. The center of the light spot (where the thermo-optic effect has the largest contribution) is first heated because of the high energy density. However, the thermal expansion of the center region is restricted (characterized by the smallest modification factor) by the outer layers. Therefore, the cavity resonance undergoes a blue shift induced by the negative thermo-optical effect and then cross the input laser frequency at the end of the stage I. (ii) At stage II, when the cavity is further heated and the heat is conducted from the inner layer to outer layers, the thermal expansion of the outer layers which have larger modification factors begin to respond. As
**FIG. 3.** (a) Calculated transmission of the PDMS microcavity. (b) Colored curves: the temperature variation of different layers (0th to 10th layer from top down). Black curve: the frequency detuning between the input laser and the cavity resonance. (c) Simulated result of the temperature distribution in the cross section of microsphere at the beginning of the four stages marked in Figs. 3(a) and 3(b). The two black arcs are the inner and outer boundaries of the PDMS layer. (d) The schematic illustration of the shift of the cavity resonance (blue Lorentzian peak) with respect to the fixed input laser frequency (red line) during the four stages marked in Figs. 3(a) and 3(b).

As a result, the cavity resonance is red shifted and then cross the input laser frequency in the opposite direction finally. (iii) At stage II, as the red detuning increases, the optical energy cannot be coupled into the cavity anymore and the cavity is stopped from being heated. The inner layers then get to cool down first and later the outer layers. The thermal expansion effect thus overwhelms the thermo-optic effect and pulls the cavity resonance totally below the laser frequency. (iv) At stage IV, the cavity without input power begins to cool down and approaches the input laser frequency until another cycle starts. In order to illustrate the process more intuitively, in Fig. 3(c) we show the temperature distribution of the cavity at the beginning of each stage, obtained by FEM simulation. As can be seen from Fig. 3(c), the distribution of the temperature variation shows a very good correspondence with the field energy density in Fig. 1(c), which means that the temperature of the region with larger field energy density is higher. This further confirms that this thickly-coated hybrid microcavity can indeed be regarded as a pure PDMS microcavity, and verifies the feasibility of our layered model for the PDMS microcavity. Since only the heat flow within the mode volume (instead of the whole cavity volume) is needed to be considered, the SSP in the pure PDMS microcavity has much shorter pulse period compared with several previous works.\[30–32\] Especially, the width of the first dip in one pulse is smaller than 10 ns, as shown in the inset of Fig. 2(b).

**IV. FURTHER EXPERIMENTS ON THE CHARACTERISTICS OF SSP**

To further study the characteristics of the SSP in the PDMS microcavity, we investigate the effect of the input power on the SSP period, with the result shown in Fig. 4(a). It can be seen that, for two different modes, the SSP period increases almost linearly with the input power, which is in accordance with the previous experimental results.\[32\] This can be explained as follows: with more intracavity power, the microcavity reaches a higher temperature in stages I and II. Therefore, it takes a longer time to cool itself down in stages III and IV. We also find that the duration of stage II almost remains unchanged, as shown in the insets of Fig. 4(a). This is because both the blue shift induced by the thermo-optic effect at stage I and the red shift caused by the thermal expansion effect at stage II are proportional to the temperature increase and thus to the input power. As a result, the time needed for the cavity resonance to recover from the blue shift during stage II nearly remains the same.

The process of SSP can be seen as an oscillation of the cavity resonance around the input laser wavelength and is a dynamic equilibrium. Once the equilibrium is reached, the SSP period is expected to be constant, independent of the input wavelength. We then study the effect of the input laser wavelength on the SSP period. We keep the input power of \(\sim 1.2 \text{ mW}\) and gradually change the bias voltage that is exerted onto the PZT of the laser. At each wavelength, we record the transmission after
FIG. 4. (a) Dependence of the SSP period on the input power for two modes (blue squares for mode 1 and black triangles for mode 2). Several transmissions of mode 1 under different input powers are given in the inset. (b) Effect of the input laser wavelength shift on the SSP period for another two different modes.

it is stabilized, as displayed in Fig. 4(b). As expected, for two different modes, the SSP periods remain relatively stable within a small range (∼0.5 µs) under different laser wavelengths.

V. CONCLUSIONS

In summary, we observe fast SSP in the transmission of PDMS microspheres under a fixed-frequency laser excitation. New competing mechanism based on the framework of self-pulsation is proposed, for the first time taking into consideration the temperature distribution within the mode volume. The properties of the SSP period are investigated in detail and confirm the validity of our theoretical model. The key factor of the fast SSP is that only the heat flow within the mode volume is needed to be considered, thus leading to a much smaller SSP period. This MHz-level SSP holds promise for applications in pulsed lasers or optical modulations.

ACKNOWLEDGMENTS

This work was supported by the 973 program (No. 2013CB328704), the NSFC (Nos. 11474011, 11222440, 11121091, and 61435001), and Beijing Natural Science Foundation Program (Grant No. 4132058). ZCL was also supported by the President’s Fund for Undergraduate Research of Peking University.

APPENDIX: TABLE I

TABLE I. Parameters used in numerical simulation.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
<th>Units</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D$</td>
<td>31.5</td>
<td>µm</td>
<td>measurement</td>
</tr>
<tr>
<td>$Q_0$</td>
<td>$1 \times 10^7$</td>
<td>-</td>
<td>measurement</td>
</tr>
<tr>
<td>$P_{in}$</td>
<td>1.2</td>
<td>mW</td>
<td>measurement</td>
</tr>
<tr>
<td>$n_0$</td>
<td>1.41</td>
<td>-</td>
<td>measurement</td>
</tr>
<tr>
<td>$dn/dT$</td>
<td>$-1 \times 10^{-4}$</td>
<td>K$^{-1}$</td>
<td>Ref. 26</td>
</tr>
<tr>
<td>$dR/dT/R$</td>
<td>$2.7 \times 10^{-4}$</td>
<td>K$^{-1}$</td>
<td>Ref. 26</td>
</tr>
<tr>
<td>$\rho$</td>
<td>965</td>
<td>Kg/m$^3$</td>
<td>Ref. 26</td>
</tr>
<tr>
<td>$C_p$</td>
<td>1300</td>
<td>J/Kg/K</td>
<td>Ref. 26</td>
</tr>
<tr>
<td>$K_h$</td>
<td>0.2</td>
<td>W/m/K</td>
<td>Ref. 26</td>
</tr>
<tr>
<td>$\alpha_{abs}$</td>
<td>3.45</td>
<td>m$^{-1}$</td>
<td>Ref. 38</td>
</tr>
</tbody>
</table>