Research article

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Controllable Kerr and Raman-Kerr frequency combs in functionalized microsphere resonators

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Abstract: Optical frequency comb (OFC) based on the whispering-gallery-mode microresonator has various potential applications in fundamental and applied areas. Once the solid microresonator is fabricated, its structure parameters are generally unchanged. Therefore, realizing the tunability of the microresonator OFC is an important precondition for many applications. In this work, we proposed and demonstrated the tunable Kerr and Raman-Kerr frequency combs using the ultrahigh-quality-factor (Q) functionalized silica microsphere resonators, which are coated with iron oxide nanoparticles on their end surfaces. The functionalized microsphere resonator possesses Q factors over 10^8 and large all-optical tunability due to the excellent photothermal performance of the iron oxide nanoparticles. We realized a Kerr frequency comb with an ultralow threshold of 0.42 mW and a comb line tuning range of 0.8 nm by feeding the control light into the hybrid microsphere resonator through its fiber stem. Furthermore, in order to broaden the comb span, we realized a Raman-Kerr frequency comb with a span of about 164 nm. Meanwhile, we also obtained a comb line tuning range of 2.67 nm for the Raman-Kerr frequency comb. This work could find potential applications in wavelength-division multiplexed coherent communications and optical frequency synthesis.

Keywords: tunable optical frequency combs; whispering gallery modes (WGMs); microsphere resonators; stimulated Raman scattering (SRS); iron oxide nanoparticles.

1 Introduction

Optical frequency comb (OFC) generation in whispering-gallery-mode (WGM) microresonators has attracted significant attention due to their high quality factors (Q) and small footprints [1–4]. The general principle for generating the OFCs in the WGM microresonators is derived from the parametric oscillation through four-wave mixing (FWM), which converts two pump photons into a pair of signal and idler photons symmetric to the pump frequency [5]. The WGM microresonator-based OFC is first realized in the silica microtoroid resonator [6]. After that, efficient microresonator OFC generation is realized using on-chip platforms [1] and fiber-coupled WGM resonators with ultrahigh- Q factors [7–11]. Silica possesses the advantage of low optical absorption and thus it is an ideal candidate for fabricating the ultrahigh- Q WGM resonator, which presents an excellent performance for OFC generation [6, 12–20]. Especially, the zero dispersion wavelength of the silica microsphere resonator can be easily designed by controlling its diameter (D), and it is thus an alternative platform for generating the ultralow-threshold OFCs [16, 21–23].

The tunability of the OFC, which can adjust a comb line at any position within the free spectral range (FSR) is a key function in practical applications, such as wavelength-division multiplexed coherent communications [24] and optical frequency synthesis [25]. The position of the comb line is mainly determined by the material refractive index (RI) and the resonator size. However, it is difficult to change the resonator size once the microresonator is fabricated. Currently, comb line tuning is realized through shifting the pump laser frequency for the microtoroid resonator [26], mechanical actuation for the microrod resonator [11], and using the microheater for the microring resonator [27]. As for the silica microsphere resonator, the tunability is a challenging issue due to its material and structure characteristics. In the previous works, it is mainly tuned through mechanical extrusion with disadvantages of mechanical interference [28]. Therefore, we proposed an all-optical tuning scheme for the silica microsphere resonator, which was embedded

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with iron oxide nanoparticles, but with low Q factors because the nanoparticles overlapped with the WGMs [29].

Here, we proposed and experimentally demonstrated tunable Kerr and Raman-Kerr frequency combs based on the ultrahigh- Q functionalized silica microsphere resonators. The hybrid microsphere resonator was fabricated by a carbon dioxide (CO_2) laser, or commercial fused splicer, and then it was coated with iron oxide nanoparticles on its end surface (for details about the device fabrication, see part S1 in the Supplementary Material). The control light was fed into the microsphere resonator through its fiber stem and then absorbed by the iron oxide nanoparticles, leading to tuning of the comb line due to the strong photothermal effect of the nanoparticles. Furthermore, as the WGMs are located near the equator of the microsphere, which are far from the iron oxide nanoparticles, there is no deterioration for the Q factor. As shown in Figure 1A, the system consists of a coupling microfiber and a functionalized microsphere resonator. The microfiber was used to couple the pump light and excite the OFC. In this work, the functionalized microsphere resonator with a Q

factor over 10^8 was fabricated and an ultralow parametric oscillation threshold of 0.42 mW was achieved. By adjusting the control power, we realized the comb line tuning of the Kerr frequency comb with a range of 0.8 nm and of the Raman-Kerr frequency comb with a range of 2.67 nm.

2 Results and discussion

Figure 1B shows the generation principle of an OFC (including degenerate FWM and nondegenerate FWM) and stimulated Raman scattering (SRS). By combining with degenerate and nondegenerate FWM processes, an OFC can be realized as the Kerr frequency comb requires phase matching in a wide wavelength range, which is dominated by the group velocity dispersion (GVD) in the resonator [5, 6, 16]. The total GVD in the silica microsphere resonator should be anomalous in the 1550 nm band, and this can be used to compensate the resonant frequency shift, which is induced by the self-phase and cross-phase modulation excited by the pump light [16]. The total GVD can be expressed by $\text{GVD} = -(\lambda/c) \times d^2 n_{\text{eff}} / d\lambda^2$, where c is the light velocity in vacuum, λ is the wavelength in vacuum, and n_{eff} is the effective RI [6].

Figure 2A is the scanning electron microscopy (SEM) image of the fabricated functionalized silica microsphere resonator with a diameter of about 248 μm . The rough area on the end surface is coated with iron oxide nanoparticles. In order to verify the existence of the iron oxide nanoparticles on the end surface, we measure both the elemental mapping and the energy dispersive X-ray (EDX) spectrum of the functionalized microsphere resonator. As shown in Figure 2B, the purple spots represent the ferrum (Fe) distribution, which means that the iron oxide nanoparticles only distribute on the end surface far from the WGM locations, and consequently this has no influence on the Q factors of the WGMs with low azimuthal mode numbers. The EDX spectrum in Figure 2C also shows that this area consists of oxygen (O), Fe, silicon (Si), and gold (Au). The presence of Au is due to the magnetron sputtering process before SEM analysis. We also measure the basic performance of the functionalized microsphere resonator, and a silica microfiber with a diameter of about 3 μm is selected to couple the pump light into the microresonator [30–33]. The inset of Figure 2C shows the Q factor measurement result, which presents an intrinsic Q factor (Q_0) of 2.1×10^8 . The ringing phenomenon is derived from the ultrahigh Q factor [34, 35].

As shown in Figure 3A, the black line denotes the calculated total GVD curve for the fundamental mode of the silica microsphere resonator with a diameter of 248 μm using the finite element method (COMSOL Multiphysics).

Figure 1: Schematic diagram of the proposed device. (A) Illustration of tunable optical frequency comb generated in the functionalized silica microsphere resonator. The pink ring represents the light propagation path and the black area denotes the ironoxide-nanoparticle-coated area. (B) General principle for OFC generation and stimulated Raman scattering. h is the Planck constant and $\nu_{p,s,i}$ are the frequencies of the pump, signal, and idler lights, respectively. $\omega_{p,s}$ and Ω_r are the frequencies of the pump light, Stokes light and optical phonon, respectively.

Figure 2: Characterization of the functionalized microsphere resonators.

(A) Scanning electron microscopic (SEM) image of the functionalized silica microsphere resonator with a diameter of about 248 μm. (B) Ferrum distribution on the end surface of the functionalized microsphere. (C) Measured energy dispersive X-ray spectrum of the coating area. The inset is the measured transmission spectrum. The red line is the theoretical fitting curve. (D) SEM image of the functionalized microsphere resonator with a diameter of about 139 μm.

We can find that the total GVD in the 1550 nm band is anomalous, and thus we fabricate the functionalized microsphere resonator with a diameter of about 248 μm. We also calculate the total GVD curves of the WGMs with different orders (for details about the GVD calculations, see part S3 in the Supplementary Material). The GVD curves in Figure S3B,C indicate that there are small dispersion differences among different-azimuthal-order WGMs. After that, we measure the parametric oscillation threshold and realize Kerr frequency comb generation using this functionalized microsphere resonator (for details about the experimental setup, see part S4 in the Supplementary Material). During the measurement process, in order to ensure the mechanical stability, the coupling microfiber always gets in touch with the microresonator. To build up the pump power in the microresonator, we finely tune the pump wavelength (λ_p) and enable it self-locked into a certain WGM. As shown in Figure 3B, λ_p is located at 1548.45 nm. When the pump power is 0.527 mW, the first equidistant lines symmetrically located at the two sides

of the pump wavelength with a spacing of three FSRs are generated through the degenerate FWM process [5].

Next, we study the dynamic Kerr frequency comb generation by adjusting the pump power. As shown in Figure 3B–F, with the increase of the pump power, cascaded FWM generates more comb lines due to the co-effect of degenerate and nondegenerate FWM [6, 36]. Meanwhile, the comb span increases with the pump power until 1.52 mW. As shown in Figure 3F, the Kerr frequency comb with a span of about 68 nm is achieved. Figure 3G is the zoom-in spectrum of Figure 3F, and we can find that the comb spacing is 2.1 nm, which is equal to the FSR of the microsphere resonator based on the expression: $\text{FSR} = \lambda^2 / 2\pi n_{\text{eff}} R$, where R is the radius of the resonator [37]. After that, the relation between the idler power and the pump power is obtained, which is shown in Figure 3H. We also find that an ultralow parametric oscillation threshold of 0.42 mW is achieved, and the idler power increases almost linearly with the pump power until 1.1 mW. The parametric oscillation threshold of the WGM microresonator can be expressed by [22, 38]:

Figure 3: Kerr frequency comb generation.

(A) Calculated total group velocity dispersion curves of the functionalized silica microsphere resonators with diameters of 248 and 139 μm , respectively. The inset is the fundamental mode field pattern of the microsphere resonator with a diameter of 248 μm . (B–F) Kerr frequency comb formation in the functionalized microsphere resonator with a diameter of 248 μm . (G) Zoom-in spectrum of Figure 3F. (H) Idler power as a function of the pump power. The inset shows the relation between the idler power and the signal power.

$$P_{\text{th}} = 1.54 \frac{\pi}{2} \left(\frac{Q_e}{2Q_l} \right) \frac{n^2 V_{\text{eff}}}{n_2 \lambda_p Q_l^2} \quad (1)$$

where Q_e and Q_l are the external-coupled and loaded Q factors, respectively, $n=1.45$ is the linear RI of silica, $n_2=2.2 \times 10^{-20} \text{ m}^2/\text{W}$ is the nonlinear RI coefficient of silica, $V_{\text{eff}}=16,000 \mu\text{m}^3$ is the fundamental-mode volume, $\lambda_p=1548.45 \text{ nm}$ is the pump wavelength. By substituting the parameters into Eq. (1), the theoretical threshold $P_{\text{th}}=0.23 \text{ mW}$ is comparable with the measured threshold. When the pump power further increases, the idler power tends to be saturated because it is converted to other comb lines due to cascaded FWM. We also obtain the relation between the signal power and the idler power as the pump power increases. As shown in the inset of Figure 3H, both

the signal and idler sidebands generate simultaneously with nearly equal powers. Therefore, it proves that these two sidebands are generated from the same pump wavelength, which is derived from the parametric interaction in the microresonator [5, 21].

After that, we tuned the comb lines by feeding the control light into the microsphere resonator through its fiber stem. The microsphere resonator is attached on the end of a single mode fiber. As shown in Figure S2, though there is no fiber core in the junction between the fiber stem and the microsphere, the control light can still propagate inside the microsphere along the axial direction of the fiber stem and towards the nanoparticle-coated area. We calculate the electric field distribution of the control light by performing a two-dimensional (2D) simulation using the

Figure 4: Controllable Kerr frequency comb.

(A–D) Kerr frequency combs for different control powers. (E) Tuning of the Kerr frequency comb line with the increase of the control power. (F) Zoom-in spectrum of Figure 4E. (G) Comb line shift (red line), comb span (black line), and comb spacing variation (the inset) vs. the control power.

finite difference time domain (FDTD) method (FDTD solutions). (For details about the simulation result, see part S2 in the Supplementary Material.) From the simulation result shown in the Supplementary Material, the control light directly propagates through the end surface and can be efficiently absorbed by the iron oxide nanoparticles. Figure 4 is the tuning performance of the Kerr frequency comb based on the functionalized microsphere resonator with a diameter of 248 μm when the pump power is fixed at 1.42 mW. Figure 4A–D denotes the corresponding OFCs for different control powers. We find that with the increase of the control power, the whole OFC spectrum has little change apart from the comb line shift. Figure 4E shows the comb line shift vs. the control power. From Figure 4F, which is the zoom-in spectrum of Figure 4E, we can find

that the comb line presents a red shift as the control power increases from 0 to 58 mW. During the tuning process, the comb spacing always maintains one FSR. Figure 4G shows the comb line shift and the comb span vs. the control power. The comb line shift increases with the control power, and a tuning range of 0.8 nm is realized when the control power increases to 58 mW, which constitutes 38% of one FSR. The resonance wavelength shift $\Delta\lambda$ as a function of the temperature variation Δt can be expressed by [39]:

$$\Delta\lambda = \lambda_0 \left(\frac{1}{n} \cdot \frac{\partial n}{\partial t} + \frac{1}{R} \cdot \frac{\partial R}{\partial t} \right) \Delta t \quad (2)$$

where λ_0 is the initial resonance wavelength, $\frac{1}{n} \cdot \frac{\partial n}{\partial t} = 8.52 \times 10^{-6} \text{ K}^{-1}$, and $\frac{1}{R} \cdot \frac{\partial R}{\partial t} = 5.5 \times 10^{-7} \text{ K}^{-1}$ [40, 41].

By substituting these parameters into Eq. (2), we calculate the temperature variation as $\Delta t = 57$ K, which leads to the RI increase of 7×10^{-4} and the radius increase of $0.0039 \mu\text{m}$. Such small parameter variations have little influence on the total GVD of the WGMs. As shown in Figure 4G, the comb span has a small variation of about 2 nm, which verifies the fact that the total GVD changes little during the tuning process. According to the FSR expression, it is evidently seen that the comb spacing tends to decrease as n_{eff} and R increase, which is derived from the increase of the control power and can be seen in the inset of Figure 4G. The maximum comb spacing variation of 141 MHz is calculated corresponding to the control power of 58 mW. Besides, according to our previous work, the response time should be millisecond level [37].

Though the microsphere resonator possesses the advantage for the ultralow-threshold OFC generation, it is difficult to realize the flattened dispersion in a wide wavelength range. Consequently, it is challenging to realize the wide-span Kerr frequency comb in the microsphere

resonator. SRS in the silica microresonator is the phenomenon that can generate the Stokes wavelength (λ_{stokes}) which is about 13 THz away from the pump wavelength [42]. Therefore, the comb span can be broadened by combining with SRS [38, 43–45]. Here, we also realize the Raman-Kerr frequency comb with a wider span compared with the Kerr frequency comb. As shown in Figure 2D, we fabricate the functionalized silica microsphere resonator with a diameter of about $139 \mu\text{m}$ and Q_0 of 3.5×10^8 . A CO_2 laser is used to fabricate the microsphere resonator with a relatively small diameter. As shown in Figure 3A, the zero dispersion wavelength of the silica microsphere resonator with a diameter of $139 \mu\text{m}$ is about 1590 nm. λ_p is fixed at 1553.7 nm, which is in the normal GVD region, and thus it is difficult to excite degenerate FWM around the pump light [46]. However, in this case, λ_{stokes} is located in the anomalous GVD region, which leads to FWM generation near λ_{stokes} [38]. As shown in Figure 5A, when the pump power is 0.191 mW, both the sidebands near λ_p and λ_{stokes} are not generated. Figure 5B shows that there are several comb lines near λ_p , which are

Figure 5: Raman-Kerr frequency comb generation.

(A–E) Raman-Kerr frequency comb formation in the functionalized microsphere resonator with a diameter of $139 \mu\text{m}$. (F) First-order sideband power and Stokes power as functions of the pump power. (G) Close-up view of Figure 5E.

Figure 6: Controllable Raman-Kerr frequency comb.

(A) Tuning of the Raman-Kerr frequency comb line with the increase of the control power. (B) (Black line frame in Figure 6A), (C) (red line frame in Figure 6A), and (D) (blue line frame in Figure 6A) Zoom-in spectra of Figure 6A. (E) Comb line shift (red line), comb span (black line), and comb spacing variation (the inset) vs. the control power.

derived from nondegenerate FWM between the pump light and Stokes comb lines, when the pump power is 0.503 mW. After that, we further increase the pump power to measure the evolution of the Raman-Kerr frequency comb. As shown in Figure 5C, when the pump power is 0.569 mW, more comb lines emerge between λ_p and λ_{Stokes} . Furthermore, as shown in Figure 5C–E, the comb span also increases with the pump power. As shown in Figure 5E, when the pump power increases to 0.954 mW, the Raman-Kerr comb with a span of about 164 nm is obtained. Figure 5G is the zoom-in spectrum of Figure 5E, and it shows that the comb spacing is 3.88 nm, which is equal to the FSR of the microsphere resonator. The first-order sideband power and the Raman power as functions of the pump power are also shown in Figure 5F. We can find that the first-order sideband power and the Raman power increase rapidly with the pump power until 0.569 mW. As the pump power further increases, both the first-order sideband power and the Raman power tend to be stable due to the consumption by degenerate and nondegenerate FWM. It is worthy to be noted that the thresholds of the first-order sideband and SRS are roughly equal, which can be verified by the aforementioned theoretical analysis.

After that, the tuning of the Raman-Kerr frequency comb is also performed. The pump power is fixed at 0.859 mW. Figure 6A shows the tunable Raman-Kerr comb spectrum when the pump power increases from 0 to 112 mW. As shown in Figure 6B, from the enlarged spectrum near the pump wavelength, we can clearly find that the comb lines present a red shift, consistently, while always maintaining the comb spacing as one FSR. From Figure 6C, we can find that all the comb lines between λ_p and λ_{Stokes} are always present with the increase of the control power. Also, from Figure 6D, almost no comb line at the right edge of the Raman-Kerr frequency comb spectrum disappears as the control power increases. Figure 6E shows the comb line shift and the comb span vs. the control power. From the red line, we can find that the comb line shift increases with the control power, and a tuning range of 2.67 nm, which constitutes 69% of one FSR, is achieved when the control power increases to 112 mW. Considering the thermo-optic coefficient and thermal expansion coefficient, this tuning range corresponds to a temperature variation of 190 K, which leads to a RI variation of 0.0023 and a radius variation of 0.0073 μm , and this means that there are little GVD changes. Furthermore,

from the black line in Figure 6E, we find that the comb span varies by only about 15.4 nm, which verifies the GVD stability during the tuning process. Besides, as shown in the inset of Figure 6E, the maximum comb spacing variation is calculated as 840 MHz.

3 Conclusion

In conclusion, we have demonstrated tunable Kerr and Raman-Kerr frequency combs based on the functionalized microsphere resonators, which are coated with the iron oxide nanoparticles on their end surfaces. We first fabricated the hybrid microsphere resonator with a Q factor over 10^8 , which possesses the large all-optical tunability. Using the hybrid microsphere resonator with a diameter of 248 μm , which has the anomalous GVD in the 1550 nm band, we obtained a Kerr frequency comb with an ultralow threshold of 0.42 mW. Due to its large tunability, we realized the tuning of the comb line within a range of 0.8 nm. Furthermore, we also realized a Raman-Kerr frequency comb with a span of 164 nm. Meanwhile, we obtained a large comb line tuning range of 2.67 nm, which constitutes 69% of one FSR. This work could find applications in wavelength-division multiplexed coherent communications and optical frequency synthesis.

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References

- [1] Gaeta AL, Lipson M, Kippenberg TJ. Photonic-chip-based frequency combs. *Nat Photonics* 2019;13:158–69.
- [2] Kippenberg TJ, Gaeta AL, Lipson M, et al. Dissipative Kerr solitons in optical microresonators. *Science* 2018;361:eaan8083.
- [3] Chembo YK. Kerr optical frequency combs: theory, applications and perspectives. *Nano Photonics* 2016;5:214–30.
- [4] Jung H, Tang HX. Aluminum nitride as nonlinear optical material for on-chip frequency comb generation and frequency conversion. *Nano Photonics* 2016;5:263–71.
- [5] Kippenberg TJ, Spillane SM, Vahala KJ. Kerr-nonlinearity optical parametric oscillation in an ultrahigh-Q toroid microcavity. *Phys Rev Lett* 2004;93:083904.
- [6] Del'Haye P, Schliesser A, Arcizet O, et al. Optical frequency comb generation from a monolithic microresonator. *Nature* 2007;450:1214–7.
- [7] Wang CY, Herr T, Del'Haye P, et al. Mid-infrared optical frequency combs at 2.5 μm based on crystalline microresonators. *Nat Commun* 2013;4:1345.
- [8] Herr T, Brasch V, Jost JD, et al. Temporal solitons in optical microresonators. *Nat Photonics* 2013;8:145–52.
- [9] Yang KY, Beha K, Cole DC, et al. Broadband dispersion-engineered microresonator on a chip. *Nat Photonics* 2016;10:316–20.
- [10] Yang QF, Yi X, Yang KY, et al. Stokes solitons in optical microcavities. *Nat Phys* 2016;13:53–7.
- [11] Papp SB, Del'Haye P, Diddams SA. Mechanical control of a microrod-resonator optical frequency comb. *Phys Rev X* 2013;3:031003.
- [12] Vahala KJ. Optical microcavities. *Nature* 2003;424:839–46.
- [13] Zhang XY, Cao QT, Wang Z, et al. Symmetry-breaking-induced nonlinear optics at a microcavity surface. *Nat Photonics* 2019;13:21–4.
- [14] Yang SC, Wang Y, Sun HD. Advances and prospects for whispering gallery mode microcavities. *Adv Opt Mater* 2015;3:1136–62.
- [15] Chen WJ, Kaya Özdemir Ş, Zhao GM, et al. Exceptional points enhance sensing in an optical microcavity. *Nature* 2017;548:192–6.
- [16] Agha IH, Okawachi Y, Foster MA, et al. Four-wave-mixing parametric oscillations in dispersion-compensated high-Q silica microspheres. *Phys Rev A* 2007;76:043837.
- [17] Yu WY, Jiang WC, Lin Q, et al. Cavity optomechanical spring sensing of single molecules. *Nat Commun* 2016;7:12311.
- [18] Yang Y, Jiang XF, Kasumie S, et al. Four-wave mixing parametric oscillation and frequency comb generation at visible wavelengths in a silica microbubble resonator. *Opt Lett* 2016;41:5266–9.
- [19] Song QH. Emerging opportunities for ultra-high Q whispering gallery mode microcavities. *Sci China Phys Mech Astron* 2019;62:074231.
- [20] Jiang XF, Shao LB, Zhang SX, et al. Chaos-assisted broadband momentum transformation in optical microresonators. *Science* 2017;358:344–7.
- [21] Castro-Beltrán R, Diep VM, Soltani S, et al. Plasmonically enhanced Kerr frequency combs. *ACS Photonics* 2017;4:2828–34.
- [22] Shen XQ, Beltran RC, Diep VM, et al. Low-threshold parametric oscillation in organically modified microcavities. *Sci Adv* 2018;4:eaao4507.
- [23] Riesen N, Afshar VS, François A, et al. Material candidates for optical frequency comb generation in microspheres. *Opt Express* 2015;23:14784–95.
- [24] Pfeifle J, Brasch V, Laueremann M, et al. Coherent terabit communications with microresonator Kerr frequency combs. *Nat Photonics* 2014;8:375.
- [25] Cundiff ST, Ye J, Hall JL. Optical frequency synthesis based on mode-locked lasers. *Rev Sci Instr* 2001;72:3749–71.
- [26] Del'Haye P, Herr T, Gavartin E, et al. Octave spanning tunable frequency comb from a microresonator. *Phys Rev Lett* 2011;107:063901.
- [27] Xue XX, Xuan Y, Wang C, et al. Thermal tuning of Kerr frequency combs in silicon nitride microring resonators. *Opt Express* 2016;24:687–98.

- [28] Dinyari KN, Barbour RJ, Golter DA, et al. Mechanical tuning of whispering gallery modes over a 0.5 THz tuning range with MHz resolution in a silica microsphere at cryogenic temperatures. *Opt Express* 2011;19:17966–72.
- [29] Zhao P, Shi L, Liu Y, et al. Iron-oxide nanoparticles embedded silica microsphere resonator exhibiting broadband all-optical wavelength tunability. *Opt Lett* 2014;39:3845–8.
- [30] Zhu S, Xiao BW, Jiang B, et al. Tunable Brillouin and Raman microlasers using hybrid microbottle resonators. *Nano Photonics* 2019;8:931–40.
- [31] Wang KY, Gu ZY, Sun WZ, et al. Quasi-guiding modes in microfibers on a high refractive Index substrate. *ACS Photonics* 2015;2:1278–83.
- [32] Zhu S, Shi L, Xiao BW, et al. All-optical tunable microlaser based on an ultrahigh-Q erbium-doped hybrid microbottle cavity. *ACS Photonics* 2018;5:3794–800.
- [33] Yu XC, Li BB, Wang P, et al. Single nanoparticle detection and sizing using a nanofiber pair in an aqueous environment. *Adv Mater* 2014;26:7462–7.
- [34] Rasoloniaina A, Huet V, Nguyễn TKN, et al. Controlling the coupling properties of active ultrahigh-Q WGM microcavities from undercoupling to selective amplification. *Sci Rep* 2014;4:4023.
- [35] Dong CH, Zou CL, Cui JM, et al. Ringing phenomenon in silica microspheres. *Chin Opt Lett* 2009;7:299–301.
- [36] Kippenberg TJ, Holzwarth R, Diddams SA. Microresonator-based optical frequency combs. *Science* 2011;332:555–9.
- [37] Zhu S, Shi L, Yuan SX, et al. All-optical control of ultrahigh-Q silica microcavities with iron oxide nanoparticles. *Opt Lett* 2017;42:5133–6.
- [38] Liu XW, Sun CZ, Xiong B, et al. Integrated high-Q crystalline AlN microresonators for broadband Kerr and Raman frequency combs. *ACS Photonics* 2018;5:1943–50.
- [39] Dong CH, He LN, Xiao YF, et al. Fabrication of high-Q polydimethylsiloxane optical microspheres for thermal sensing. *Appl Phys Lett* 2009;94:231119.
- [40] Leviton DB, Frey BJ. Temperature-dependent absolute refractive index measurements of synthetic fused silica. *Proc SPIE* 2006;6273:62732K.
- [41] White GK. Thermal expansion of reference materials: copper, silica and silicon. *J Phys D Appl Phys* 1973;6:2070–8.
- [42] Spillane S, Kippenberg T, Vahala KJ. Ultralow-threshold Raman laser using a spherical dielectric microcavity. *Nature* 2002;415:621–3.
- [43] Choi H, Armani AM. Raman-Kerr frequency combs in Zr-doped silica hybrid microresonators. *Opt Lett* 2018;43:2949–52.
- [44] Aghal H, Okawachi Y, Gaeta AL. Theoretical and experimental investigation of broadband cascaded four-wave mixing in high-Q microspheres. *Opt Express* 2009;17:16209–15.
- [45] Fujii S, Kato T, Suzuki R, et al. Transition between Kerr comb and stimulated Raman comb in a silica whispering gallery mode microcavity. *J Opt Soc Am B* 2018;35:100–6.
- [46] Li M, Wu X, Liu LY, et al. Kerr parametric oscillations and frequency comb generation from dispersion compensated silica micro-bubble resonators. *Opt Express* 2013;21:16908–13.

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