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光纤微流传感技术研究进展

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摘要：本文介绍了本课题组在光纤微流激光传感器和无源光纤微流传感器两方面的研究进展。光纤微流激光传感器利用光纤微流激光的输出变化来探测生化参数的改变。光纤截面作为环形微腔形成光反馈，增强了腔内光子和待测物质的相互作用，从而提高了微流激光的传感灵敏度。此外，光纤尺寸均匀，易低成本、批量制作光纤微腔，可制备高重复性或一次性使用的光纤微流激光。本文还介绍了基于光力/光热效应的无源光纤微流传感器。该类传感器利用光产生的力学或热学效应对微流体进行温度、流速、浓度传感，具有灵活性高、集成度好、多功能、可重构等特点。

关键词：光纤微流传感；光纤微流激光；光纤传感；光力效应；光热效应

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Recent advances in fiber optofluidic sensors

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Abstract: In this mini-review, recent advances in the fiber optofluidic lasers and passive fiber optofluidic sensors are introduced. Fiber optofluidic laser can detect the biochemical changes using its laser output as a sensing signal. The cross-section of fiber can be used as a microcavity, providing optical feedback. The microcavity enhances the light-matter interaction, thus increasing the sensitivity. Furthermore, the geometry of optical fibers is uniform, easy to be mass produced with low cost, can be used to realize highly reproducible and disposable optofluidic laser. Passive fiber optofluidic sensors are also introduced based on the laser induced force and photo-thermal effects, which is flexible, easy to be integrated, multi-functional and reconfigurable.

Keywords: fiber optofluidics sensor; fiber optofluidics laser; fiber sensing; laser induced force; photo-thermal effect

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1 引　言

光微流(Optofluidics)将光子技术与微流结合，可形成多功能、集成化的微系统。微流具有固体材料无法比拟的独特特性：1) 在两种不相容的液体界面具有天然的光滑特性；2) 可通过扩散实现渐变折射率分布；3) 可以方便地流动和控制；4) 可通过液体混合实现密度调节或掺杂。光子技术可用于检测、操控微

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流及其中的微纳样品，进而实现光微流传感技术[1-2]。

光纤集成了导光和微流通道功能，正受到研究者的广泛关注，形成了新型光纤微流传感技术。在光纤操控及其传感方面，李宝军课题组[3]利用亚波长量级的微光纤在液体中实现了胶状微球的分选；苑立波课题组[4]利用光纤实现光阱，可捕获微球，进一步实现了温度传感；Bykov 等人[2]在空心光子晶体光纤中捕获微球，并利用透射光信号和多普勒测速法分别实现了具有高空间分辨率的电场和温度传感。在光纤微流激光方面，Wang[5]、Liu[6]等人采用光纤制成的回音壁(whisper gallery mode, WGM)微腔实现了光纤微流激光输出；Zhang[7]等人采用具有光子带隙(photonic band-gap, PBG)的光纤作为反馈腔，实现了径向的微流激光输出。王璞团队[8]采用空心无节抗谐振光纤(hollow-core nodeless anti-resonant fiber, HARF)测量了样品的拉曼光谱。Gu 等人[9]通过调节空间泵浦实现了光纤上聚合物微瓶谐振腔的单模 WGM 激光。Gerosa 等人[10]实现了全光纤高重频输出的光纤微流激光。

围绕光纤在微流传感中的优势，本文分别介绍光纤微流激光传感器和无源光纤微流传感器。其中，光纤微流激光器用光纤作为微谐振腔实现激光输出。利用光纤结构多样性可以实现不同类型的光微流激光器。由于光纤的尺寸重复性好，这种光纤的微流激光器有望实现低成本、一次性传感。而无源光纤微流传感器以光纤将激光导入微流体，利用光照射液体产生的物理效应实现传感。

2　光纤微流激光传感器

2.1　光微流激光传感技术

光微流激光器[11-13]包括了传统激光器三要素：谐振腔、增益介质、泵浦源。在泵浦光作用下，增益介质被激活实现粒子数反转。增益介质产生的光子在谐振腔作用下反馈回增益介质，得到进一步放大。在满足激光阈值条件后实现激光输出。谐振腔的反馈增强了光子和内部增益介质的相互作用[13]，这使得光微流激光对激光腔内部生化分子的状态非常敏感。因此，光微流激光很适合用于实现高灵敏度的微流传感器。

我们提出了将酶促反应与光微流激光器结合，实现了酶催化的光微流激光器并实现了高性能的离子传感[14]。如图 1 所示，两块宽带高反镜平行放置构成法珀腔(Fabry–Pérot cavity, FP)，位于两块高反镜中间的方形毛细管充当微流通道实现液体流入流出。利用免

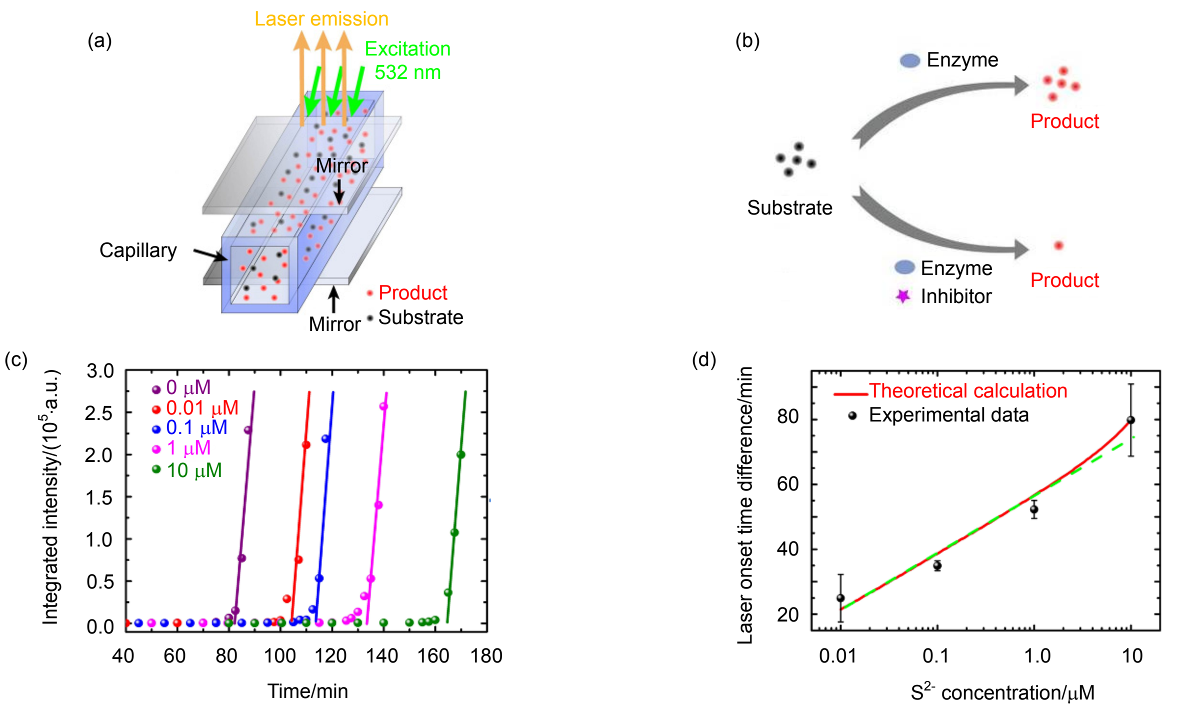

图 1　光微流激光用于 S^{2-}传感。(a) 光微流激光离子传感器结构示意图；(b) 酶催化反应及抑制剂作用示意图；(c) 激光输出强度与时间的关系曲线；(d) 不同 S^{2-}浓度下激光出射时间

Fig. 1　S^{2-} detection based on optofluidic laser. (a) Structure of the laser cavity for the optofluidic catalytic laser; (b) Generation of the product as gain material and effect of the inhibitor on the catalytic reaction; (c) Spectrally integrated intensity as a function of reaction time with different S^{2-} concentrations; (d) Laser onset time difference versus S^{2-} concentration

疫检测中广泛使用的辣根过氧化物酶，将无色底物催化为荧光产物。在泵浦光作用下，该荧光产物可作为激光增益介质。随着酶促反应的进行，荧光产物的含量逐渐升高，当产物浓度满足激光阈值条件可实现激光输出。当反应体系中有酶抑制剂如 S^{2-}存在时，酶的活性降低，荧光产物浓度增加速率放缓，使得激光出射时间延长(如图 1(c)所示)。激光出射时间和反应体系中 S^{2-}浓度相关。因此，通过测量激光出射时间，可实现 S^{2-}浓度传感(如图 1(d)所示)。最终，该方法实现了高性能 S^{2-}传感，探测极限为 10 nM，动态范围为 3 个量级。

另外，结合新型发光材料，可实现具有新的传感特性的光微流激光[15]。

2.2 新型光纤微流激光器及传感技术

一次性传感器具有本质安全、成本低、重复性高等优点，在生化检测中具有广泛应用[16]。光微流激光具有高灵敏度，但之前的光微流激光器无法满足一次性的检测需求。实现一次性的光微流激光器的关键在于解决如何高重复、低成本、大批量地制作微腔。光纤由于在其拉制过程中，尺寸得到了很精确的控制，在作为激光谐振腔时，具有很好的重复性。以康宁公司 SMF-28e 为例，其每千米的尺寸波动率大约为 0.56%。同时，光纤也可以低成本、大批量地制作。普通商用光纤每米的成本约为 0.01 美元，超低成本是大规模应用的基础之一。图 2 展示了利用微结构光纤实现的高重复光纤微流激光器[17]。微结构光纤穿过两根玻璃毛细管，其空间位置被玻璃毛细管限制。如图 2(b)所示，光纤外壁的 WGM 可通过倏逝场和内部增益介质相互作用。实验证实，不同光纤之间的激光输出强度波动约为 6.5%(如图 2(c)所示)，足以满足生化传感的应用。

高通量检测是生化传感中的又一重要需求[18]。光纤通过集成阵列的方式有望实现高通量的传感。如图 2(d)所示，微结构光纤通过 V 型槽平行排布，调节各个通道光纤使其取向一致，可以克服光纤微结构空间不对称性引起的激光方向性。532 nm 的脉冲泵浦光经过球面透镜汇聚于光纤阵列，光纤阵列的位置可通过

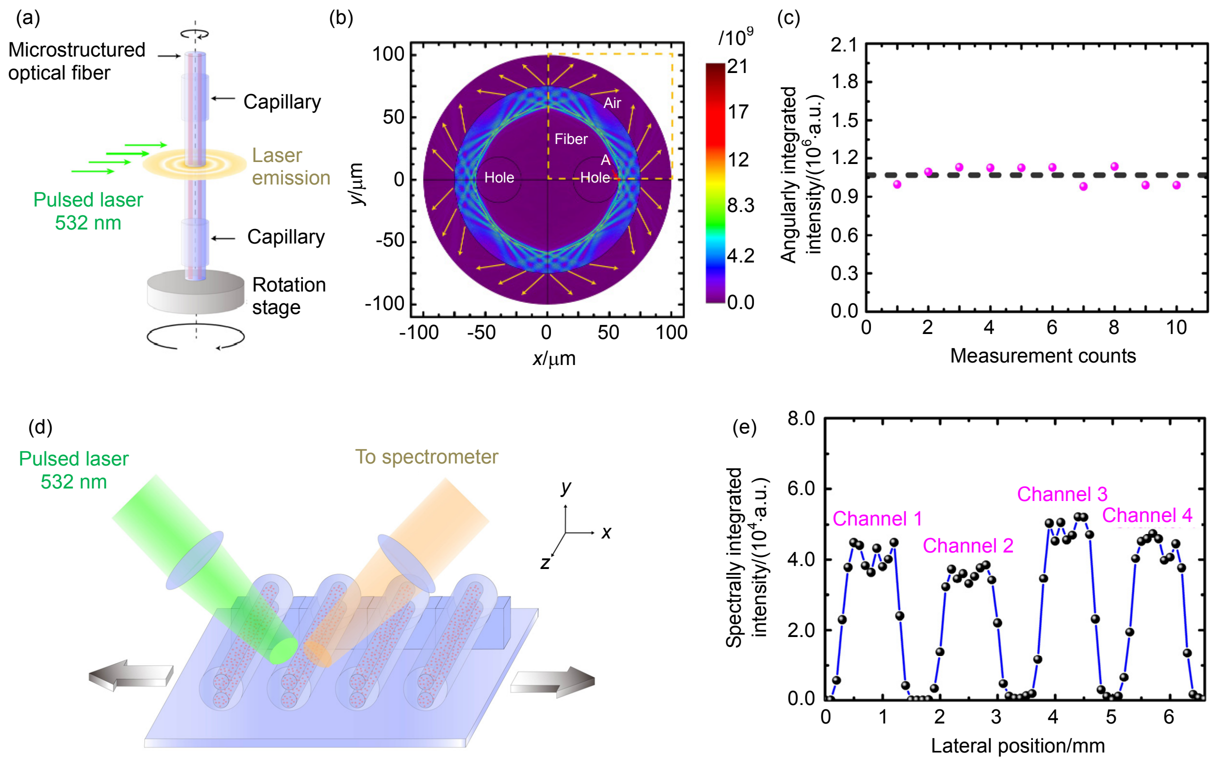

图 2 高重复光纤微流激光器[17]。(a) 高重复性光纤微流激光器实验装置图；(b) 微结构光纤横截面光场分布仿真结果；(c) 微结构光纤输出重复性实验结果；(d) 光纤微流激光阵列示意图；(e) 光纤微流激光各通道输出强度

Fig. 2 Reproducible fiber optofluidic laser[17]. (a) Schematic diagram of the experimental setup for fiber optofluidic laser; (b) Intensity distribution in the cross-section of the MOF; (c) Angular integrated intensity using 10 sections of MOFs; (d) Schematic diagram of the FOFL array; (e) The spectrally integrated intensity as a function of the lateral pump position

电动位移台在 x 方向精细调节，从而实现泵浦光对各通道的扫描，各通道发射的激光被光纤收集并送入光谱仪，实现了通道间的强度的高重复性(如图 2(e)所示)[17]。此外，利用单纵模光纤微流激光实现波分复用也是解决高通量检测的有效途径，我们利用微结构光纤的滤波效应，实现了单纵模的光纤微流激光输出[19]，信噪比达 21 dB，单纵模线宽 53 pm。在 20 nm 波长范围内，该方式理论上可以实现近 400 个通道的复用。

另外，Chen 等人[20]通过化学交联的方式在光纤外壁制作了一层单分子增益薄膜，利用光纤外壁的 WGM 提供光反馈证实了单分子层增益介质也能实现激光输出，使得超低样品用量的光微流激光器成为可能。Lee 等人[21]将单分子层激光应用于 DNA 检测，具有低荧光背景、超低样品用量的特点。

3　无源光纤微流传感器

光纤因其优良的波导特性和小巧体积，可伸入微流通道内部；利用光物理效应，如光力效应[22-26]和光热效应[27-31]，可以在微流通道内部实现对液体及其溶质的高性能传感。光纤微流传感器具有灵活性高、集成度好、多功能、可重构等特点。

3.1　基于光力效应的光纤微流传感器

结合微流系统提供的流体力，基于光力效应的光纤微流系统实现了可调操控[24-26]和传感[27-28]。这种光纤微流技术的工作原理如图 3(a)所示[27]。激光沿光纤传输，从光纤端面出射并照射在微粒上，由于辐射压和光子动量传递，对微粒产生光力效应。垂直光纤的平面上，光力分量表示为梯度力(F_{tg})，方向指向光场密度最大处，大小从光轴向外逐渐减小，起到将微粒限制在光轴上的作用。平行于光纤的平面上，光力的分量表示为散射力(F_{ao})，方向沿光传输方向，大小沿光传输方向逐渐减小，起到将微粒推离光纤端面的作用。微流系统在与光传输方向相反的方向提供一个流体力(F_v)，与光散射力 F_{ao} 平衡，实现对微粒的捕获。通过调节流速(v)的大小改变 F_v，可改变 F_{ao} 与 F_v 的平衡位置，进而改变微粒的捕获位置，实现对微粒可调操控。基于上述原理，我们利用平端面单模光纤，实现了测量范围在 20 nL/min~22 μL/min 的流速传感[26]。同时，该装置还可实现最大操控距离为 715 μm 的可调光操控。流速传感实验结果如图 3(b)所示。

上述基于微粒操控位置探测的光纤流速传感器，其传感性能受到操控距离的限制：对于高流速探测，由于微粒被操控到距离光纤很近的位置，传感灵敏度降低，使其达到探测极限。针对这一问题，我们提出了一种双模式的光纤微流传感系统[28]。该系统在开环模式下，与上述系统原理相同，利用微粒的操控距离来标定流速，结构简单，在低流速区域具有很高的灵敏度(图 4(a))；在闭环模式下，当流速变化时，通过主动调节激光器输出功率，使得光力大小发生改变并与流体力平衡，利用反馈控制将微粒固定在原来的位置。因此，激光器的输出功率与流速具有线性关系(图 4(b))。该流速传感器两种模式可自由切换，实现了从 10 nL/min~100000 nL/min 的大动态范围流速传感(图 4(c))[28]。

3.2　基于光热效应的光纤微流传感器

基于光热效应，可实现光纤微流速传感器[29-30]。当光照射到液体中，由于光热效应，光能转化为液体的热能，使局部液体温度升高，造成液体蒸发或溶解

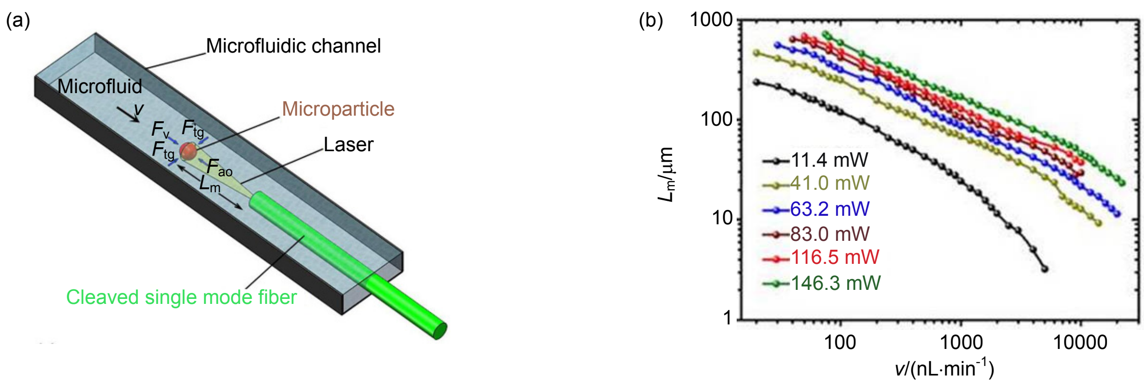

图 3　基于光力操控的流速传感[27]。(a) 基于光力操控的流速传感原理示意图；(b) 不同激光功率下流速与操控距离的关系曲线

Fig. 3　The flow rate sensor based on the optofluidic manipulation[27]. (a) Principle for flow rate detection; (b) Manipulation length versus flow rate at different laser powers

气体逸出，可产生气泡。为有效降低产生气泡的激光功率阈值，形成规则的、可探测的气泡，我们通过在光纤表面沉积碳纳米管薄膜的方式，实现了基于光热效应的光纤微气泡传感器[30]，其装置如图 5(a)所示。当 980 nm 连续光照射到碳纳米管薄膜上时，由于碳纳米管特殊的热学性质，可使光纤端面在较低的功率下产生规则的气泡。气泡前后反射面形成法珀腔。气泡直径随激光照射时间变化情况如图 5(b)所示。通过检测气泡的生长速率，该装置可对液体环境(如温度、流速)改变的探测。通过探测微气泡法珀腔的干涉光谱，该装置实现了 25 ℃~45 ℃的温度传感，以及 0~150 nL/min 的低流速传感[30]。

由于碳纳米管薄膜通过光力沉积，多次使用后会引起薄膜脱落，使传感器性能下降，降低使用寿命。通过在光纤端面离子溅射纳米金膜，制备结构更加稳定、重复性好的光纤微气泡微流传感器[29]。该装置采用图像法进行探测，方法更简单，成本更低。研究人员利用该装置实现了质量比在 0.5%~50%范围内的蔗

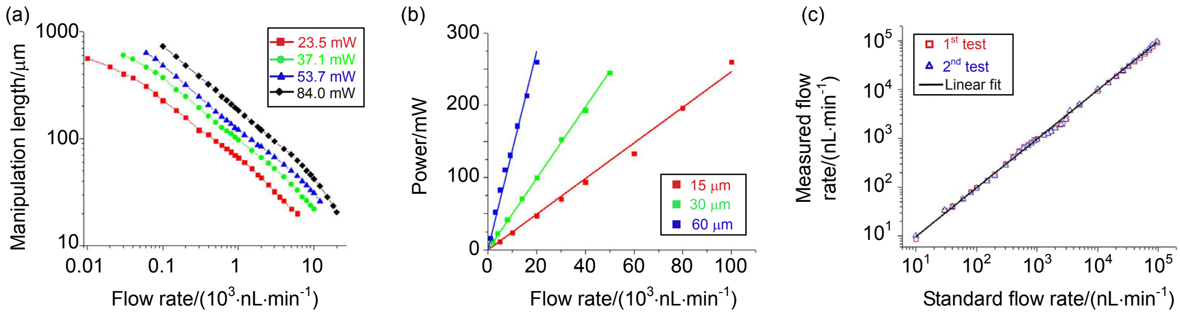

图 4　双模式流速传感。(a) 开环模式下的流速校准曲线；(b) 闭环模式下的流速校准曲线；(c) 流速传感性能曲线

Fig. 4　Dual-mode flow rate sensing. (a) Calibration of the optofluidic flow rate sensor in open-loop mode with *y* axis in log scale; (b) Calibration of the optofluidic flow rate sensor in the closed-loop mode with manipulation length fixed at 15 μm, 30 μm and 60 μm, respectively; (c) Sensing performance of the optofluidic flow rate sensor

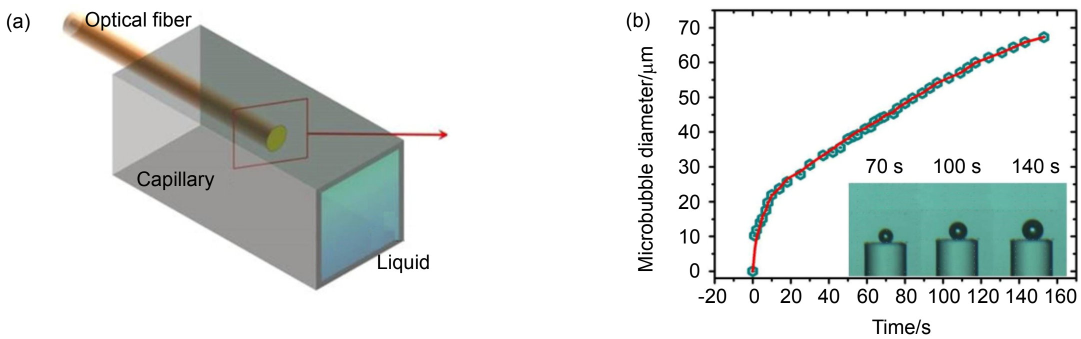

图 5　基于光热效应的光纤微流传感器。(a) 装置图；(b) 气泡腔直径变化情况

Fig. 5　The flow rate sensor based on photo thermal effect. (a) The experimental setup; (b) The generation of the fiber optofluidic microbubble-on-tip for 150 s

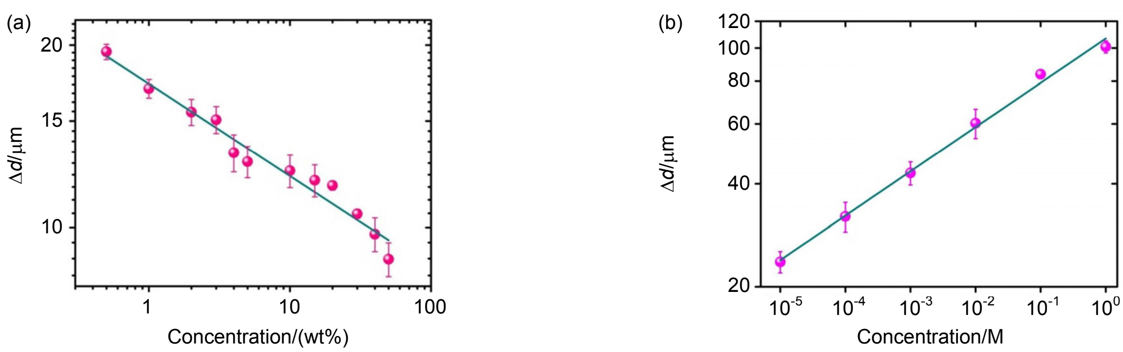

图 6　基于纳米金膜的光纤微流传感器传感性能。(a) 蔗糖浓度传感；(b) 双氧水浓度传感

Fig. 6　Concentration sensing of the fiber optofluidic sensor coated with gold nanofilm. (a) Sucrose; (b) H_2O_2

糖浓度传感(图 6(a))以及 10 μM~1 M 的大动态范围双氧水浓度传感(图 6(b))[31]。

4 结　论

本文综述了近期光纤微流传感技术的研究进展。光纤微流激光是一种新型普适性的高灵敏生化传感平台技术，可广泛用于体溶液或表面传感，所需样本体积超低、灵敏度比传统荧光方法有数量级的提升，近期发展非常迅速，并逐渐用于免疫诊断、DNA 检测等。基于光力的光纤微流传感技术，利用皮牛量级的微小光力，可实现高灵敏微流速传感；基于光热效应，可利用光热传导、微气泡产生作为传感原理，实现温度、流速、浓度等参数传感。与微流体结合，扩展了光纤传感的外延，有望发展出新型光纤生化传感手段。

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Recent advances in fiber optofluidic sensors

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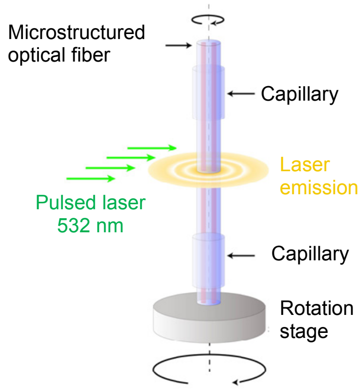

Schematic diagram of the fiber optofluidic laser

Overview: In this review, recent advances in optofluidic laser sensor and fiber optofluidic laser, as well as the passive fiber optofluidic sensors based on the optical force or photothermal effects are introduced.

Optofluidic laser (OFL) is an emerging technology that has been extensively investigated for biochemical detection. Due to the enhanced light-matter interaction, high sensitivity of OFL sensors have been demonstrated. We recently demonstrated a highly sensitive ion detection method using optofluidic laser based on Fabry-Perot cavity. A catalytic reaction that could be inhibited by the S^{2-} ion was employed to produce a fluorescence gain material for optofluidic laser. The limit of detection by the OFL method was orders of magnitude lower than the fluorescence method.

Various types of microcavities including Fabry–Perot cavity, micro ring cavity and distributed feedback schemes have been investigated for optofluidic lasing. The lasing output is highly dependent on these microcavities. The mass productions with high repeatability are difficult for previous microcavities, making it hard to realize reproducible optofluidic laser. We introduced a novel fiber optofluidic laser with high reproducible microcavities. The optical fiber can be used as a ring resonator, providing optical feedback in the cross-section for lasing. Most importantly, thanks to the precise control of the fiber geometry by draw tower, the properties (including geometry, surface properties and thus Q-factor) of microcavities along the optical fiber are almost identical. The optical fiber can be mass produced with low cost and can be utilized to realize highly reproducible and disposable optofluidic laser.

Besides the fiber optofluidic laser, passive fiber optofluidic sensors based on the laser induced force and photo-thermal effects are introduced. The laser beam offers optical force at pico-Newton scale that is very sensitive to the ambient environments. By integrating the optical fiber with microfluidic chip, single microparticle can be trapped and high performance microfluidic flow rate detection was performed based on the force balance on the microparticle. Tunable optical manipulation of microparticle was also demonstrated.

Photo-thermal effect was also introduced by optical fiber into the microfluidic chip for sensing applications. Material with high absorption, including carbon nanotube or gold nanofilm, was coated on the fiber endface. Laser absorption near the fiber tip leads to a temperature rise. Thus microbubble was generated on the fiber tip based on the photo-thermal effect. By monitoring the generation and growth of microbubble, microfluidic parameters including flow rate, temperature, and concentration can be measured. The passive fiber optofluidic sensors have the advantages of flexible, easy to be integrated, multi-functional and reconfigurable.

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