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Single-beam optical trap-based surface-enhanced Raman scattering optofluidic molecular fingerprint spectroscopy detection system

Ning Sun, Yuan Gan, Yujie Wu, Xing Wang, Shen Shen, Yong Zhu and Jie Zhang*

In this study, we developed a single-beam optical trap-based surface-enhanced Raman scattering (SERS) optofluidic molecular fingerprint spectroscopy detection system. This system utilizes a single-beam optical trap to concentrate free silver nanoparticles (AgNPs) within an optofluidic chip, significantly enhancing SERS performance. We investigated the optical field distribution characteristics within the tapered fiber using COMSOL simulation software and established a MATLAB simulation model to validate the single-beam optical trap's effectiveness in capturing AgNPs, demonstrating the theoretical feasibility of our approach. To verify the particle capture efficacy of the system, we experimentally controlled the optical trap's on-off state to manage the capture and release of particles precisely. The experimental results indicated that the Raman signal intensity in the capture state was significantly higher than in the non-capture state, confirming that the single-beam optical trap effectively enhances the SERS detection capability of the optofluidic detection system. Furthermore, we employed Raman mapping techniques to investigate the impact of the capture area on the SERS effect, revealing that the spectral intensity of molecular fingerprints in the laser-trapping region is significantly improved. We successfully detected the Raman spectrum of crystal violet at a concentration of 10^{-9} mol/L and pesticide thiram at a concentration of 10^{-5} mol/L, further demonstrating the ability of the single-beam optical trap in enhancing the molecular fingerprint spectrum identification capability of the SERS optofluidic chips. The optical trapping SERS optofluidic detection system developed in this study, as a key component of an integrated optoelectronic sensing system, holds the potential for integration with portable high-power lasers and high-performance Raman spectrometers. This integration is expected to advance highly integrated technologies and significantly enhance the overall performance and portability of optoelectronic sensing systems.

Keywords: surface-enhanced Raman scattering; optofluidic; single-beam optical trap; tapered optical fibers

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Introduction

Raman spectroscopy, due to its ability to identify unique molecular fingerprints, holds significant potential across

various fields, including chemistry, biology, and medicine. However, its application in high-sensitivity detection is limited by the inherently weak signal intensity.

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To overcome this limitation, researchers have explored various signal enhancement techniques. Among these, surface-enhanced Raman scattering (SERS) has emerged as a major focus due to its remarkable signal amplification capabilities. Surface-enhanced Raman scattering technology, with its unique electromagnetic and chemical enhancement mechanisms, enables significant amplification of Raman scattering signals. The electromagnetic enhancement arises primarily from the localized surface plasmon resonance effect, which generates intense localized electric fields on the surface of metal nanoparticles, substantially boosting the Raman signals of scattering molecules. Meanwhile, the chemical enhancement mechanism involves charge transfer processes between molecules and the metal surface, altering the electronic structure of the molecules and further enhancing the sensitivity of Raman scattering^{1–6}. Compared to other enhanced molecular spectroscopy methods, SERS offers distinct advantages, particularly its strong compatibility with diverse applications^{7–12}. The integration of SERS technology with lab-on-a-chip (LOC) systems allows for precise control of fluid movement within microchannels, enabling the execution of complex experimental procedures on a compact chip. Within these microchannels, SERS particles can effectively interact with target molecules, facilitating efficient detection. This integration not only enhances portability but also enables rapid and accurate on-site analysis, which is critical for applications in environmental monitoring, medical diagnostics, and food safety^{13–16}. Currently, optofluidic SERS detection chips can be classified into two types: those utilizing fixed metallic nanostructures and those employing non-fixed structures. Fixed nanostructures enhance the Raman signal by anchoring metallic nanostructures at specific sites within the microfluidic channels, ensuring stability in the fluid environment and producing consistent SERS effects. However, the primary limitation of this approach lies in the difficulty of replacing or adjusting the metallic nanostructures. Once contaminated or oxidized, the entire chip becomes ineffective. In contrast, non-fixed chips introduce nanoparticles alongside analytes into the microfluidic system. This method allows for easy replacement of metallic nanostructures, as the injection of fresh nanoparticles and analytes mitigates contamination or oxidation issues. However, since the nanostructures are not immobilized during fluid flow, their uneven distribution within the microchannels can reduce SERS performance and compromise the chip's

overall stability. To address these challenges, researchers have proposed several solutions, including electrochemical deposition to construct and remove metallic nanostructures on-site, the use of soluble or degradable substrate materials to simplify substrate removal and replacement, and the application of optical tweezer technology to trap metallic nanoparticles within the microchannels, thereby enhancing the performance of non-fixed chips^{17–31}. The reported optical tweezers-enhanced optofluidic SERS technology integrates optical tweezers with optofluidic SERS methodologies. This innovative approach employs the radiation pressure of a single-beam optical trap to precisely capture metal nanoparticles in the microfluidic channels, promoting their aggregation around the optical tweezers. This configuration significantly enhances the local electromagnetic field, improving both the intensity and uniformity of the Raman scattering signal. By synergistically combining optical tweezers, microfluidics, and SERS enhancement techniques, this system establishes a controllable, switch-like mechanism for amplifying molecular fingerprints^{32–41}. Currently, the application of traditional liquid Raman spectroscopy technology faces several challenges, including difficulty in achieving precise positional control, cumbersome operational procedures, and insufficient signal intensity. To overcome these challenges, the integration of SERS, optical tweezers and optical microfluidic chip technology is a good potential method, utilizing the efficient mixing and high-throughput characteristics of optical microfluidic chips.

We have developed an optical tweezer-enhanced SERS optofluidic detection system, which employs a single-beam optical trap to aggregate free silver nanoparticles (AgNPs) within an optofluidic chip, thereby enhancing SERS performance. The system integrates a 1550 nm laser module for constructing the optical trap and a 532 nm laser module for Raman signal detection. In this miniature, controllable fluidic environment, the 1550 nm laser, coupled through a tapered optical fiber, introduces optical radiation pressure into the detection chamber of the optofluidic chip. The unique design of the tapered fiber enables it to focus the laser beam, creating a high-gradient optical field at the fiber's tip. This high-gradient optical field generates optical forces that attract AgNPs suspended in the microfluidic chip, causing them to aggregate near the tapered fiber tip. As the particles aggregate, the inter-particle spacing decreases significantly. This configuration enables precise capture and

manipulation of AgNPs, reducing their separation and enhancing local surface plasmon resonance, thereby significantly amplifying the Raman signal and enabling high-sensitivity molecular fingerprint detection. The core innovation of this study lies in the development of a stable and controllable hotspot detection platform, optimized for two application scenarios. First, under laboratory conditions, the platform offers a stable, reliable, and sensitive environment for Raman detection of liquid samples. Additionally, the proposed platform seamlessly integrates with portable Raman spectrometers, forming a compact SERS detection system equipped with an integrated portable Raman detection unit. The entire system can be conveniently housed in a laboratory kit or mobile device, enabling users to independently perform liquid SERS detection tasks after brief training.

Experimental and mechanism

Figure 1(a) illustrates the structure of the single-beam optical trapping-enhanced SERS optofluidic detection system. This system comprises the optical trapping module, the optofluidic module, and the spectral detection module. For detailed information about the spectroscopic detection module and experimental apparatus, please refer to Supplementary information Section 1.

Single-beam optical trapping module

The laser signal is output through a tail fiber, ensuring efficient transmission of the optical signal. We fabricated a tapered optical fiber with a tip approximately 25 micrometers in diameter, an core angle of 0.05°, with the fabrication process and schematic provided in Supplementary information S2. The laser and tapered fiber

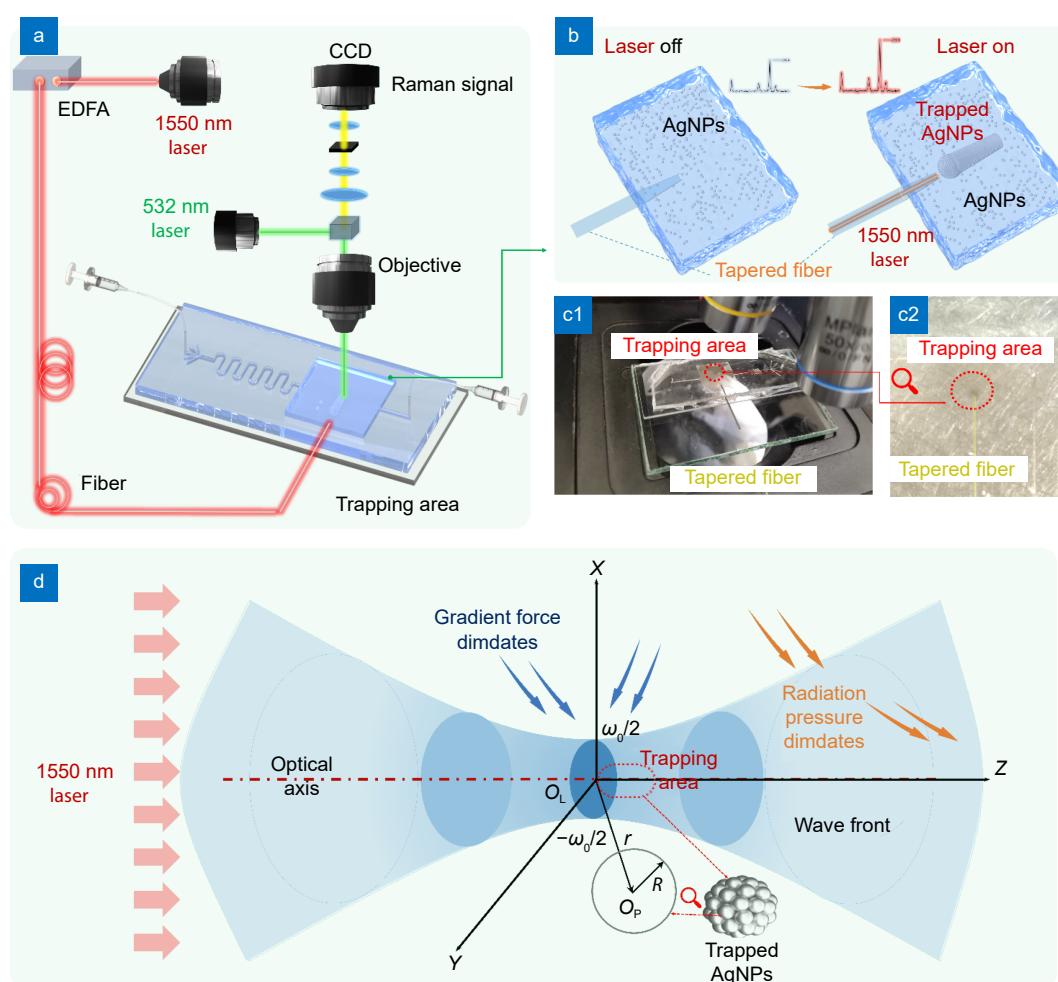


Fig. 1 | (a) Schematic diagram of the optical trapping-enhanced SERS optofluidic detection system. (b) Effect of the single-beam optical trap module switch state on AgNPs aggregation. (c1) and (c2) Operational images of the single-beam optical trap-enhanced SERS optofluidic detection system, demonstrating particle trapping and Raman detection, with (c2) providing an enlarged view of the trapping area in (c1). (d) Schematic diagram of the optical trapping mechanism.

probe are connected via a jumper, guiding the laser signal to the detection area of the microfluidic chip. As shown in Fig. 1(b), when the 1550 nm laser is activated, a single-beam optical trap is formed, capable of capturing AgNPs and concentrating them into a high-intensity SERS enhancement area. When the laser beam is turned off, the AgNPs diffuse freely again. For the detection of molecules at higher concentrations, molecular information can be obtained without 1550 nm laser irradiation (i.e., when the optical trap is in the OFF state). However, for trace molecule detection, acquiring molecular information is not possible without the focusing effect of the laser. By switching to the ON state, where light focusing is applied, the system can effectively detect and identify molecular information. For example, in applications involving water quality analysis, the ON state enables the detection of trace pollutants, thereby reducing the likelihood of missed detections.

Optofluidic module

We fabricated the microfluidic channel template using soft lithography and produced polydimethylsiloxane (PDMS) microchannels via a straightforward molding technique. Detailed fabrication methods are provided in Supplementary information S3. This module is designed for precise fluid control and molecular interaction analysis, facilitating efficient mixing of analyte molecules with AgNPs, accurate detection, and ease of operation. The preparation methods and relevant parameters for the AgNPs utilized in the experiments are detailed in Supplementary information S4. Figure 1(c1) and 1(c2) illustrate the operational setup of the single-beam optical trapping-enhanced SERS microfluidic detection system, highlighting Raman detection experiments where AgNPs are captured by the optical trap. Specifically, Fig. 1(c2) provides an enlarged view of the trapping area depicted in Fig. 1(c1). To mitigate the effects of dynamic fluctuations during Raman testing, we optimized the optofluidic environment by integrating a tapered optical fiber into the system. The experimental setup centers around the optical microfluidic chip, with the tapered optical fiber serving as a key component. The optical microfluidic system employs a high-throughput design, enabling the efficient capture and detection of even trace amounts of analytes while significantly reducing interference from the surrounding aqueous medium. The tapered optical fiber is securely anchored at the base of the chip, substantially minimizing vibrations caused by fiber movement, thereby enhancing the overall stability of the

system. During the experimentation process, a stabilization period of 3 to 5 minutes is necessary when transitioning between system states to allow the system to reach equilibrium. This interval ensures that the tapered optical fiber and the trapped AgNPs settle into their positions, thereby maintaining signal stability throughout the detection process.

Mechanism of single-beam optical trap-based SERS

In 1986, Ashkin A. demonstrated the use of a highly focused laser beam to generate a gradient force, enabling microparticles to reach equilibrium at specific positions along the optical axis, thus achieving three-dimensional trapping of microparticles. A single-beam trapping technique, relying on a single laser beam, is referred to as single-beam optical gradient trapping⁴². The high-gradient optical field created by the focused lens ensures that when the axial gradient force exceeds the scattering force, microparticles are trapped along the beam direction, forming a three-dimensional optical potential well. Particles near the focal point are stably confined within this optical trap, and any deviation induces a restoring force, maintaining stable confinement in the three-dimensional space. This phenomenon occurs because when a laser beam illuminates an object's surface, the beam's momentum is transferred to the object, generating a mechanical effect known as radiation pressure or optical pressure. This mechanical effect results from the momentum transfer of the laser beam. When the laser is focused by a lens, it creates an optical potential well at the focal point, which exerts mechanical forces on small objects. These forces can be divided into scattering forces and gradient forces. As illustrated in Fig. 1(d), the scattering force arises from the reflection and refraction of the laser beam by the particles, driving them along the beam's propagation direction. The gradient force, in contrast, is due to the uneven spatial distribution of light intensity, trapping the particles at the position where the intensity gradient is strongest. Optical tweezers primarily rely on the gradient force, often termed the "trapping force", to confine particles. Methods for calculating the gradient and scattering forces in the trapping of metallic nanoparticles by optical tweezers are provided in Supplementary information S5. The tapered optical fiber in our system generates an optical field with a high convergence angle, largely due to the inclined surface of the fiber, which facilitates a larger beam convergence angle. However, this inclined surface also introduces a mode leakage

effect, causing some light to be lost during refraction and reflection, resulting in a specific optical field distribution. This optical field creates a small region at the tip of the tapered fiber where the field intensity reaches its maximum. This design enables precise capture of AgNPs within the high-intensity optical field, significantly reducing the inter-particle spacing. SERS technology enhances molecular Raman fingerprint spectra by leveraging the surface plasmon resonance of AgNPs under incident light excitation. The enhancement effect becomes more pronounced as the distance between the AgNPs decreases. In our experiments, we used AgNPs with a diameter of approximately 50 nm. Given that the radius of these AgNPs is much smaller than the wavelength of the 1550 nm incident light typically used in optical tweezers, the Rayleigh scattering approximation is applicable within the electromagnetic wave model. The reason for the wavelength selection in optical tweezers is explained in section S6 of the SI. Under this approximation, the AgNPs can be treated as point dipoles.

The propagation of the Gaussian beam from the tapered optical fiber in water can be described by the following formula:

$$\omega(z) = \omega_0 \sqrt{1 + \left(\frac{z}{z_R}\right)^2}, \quad (1)$$

where $\omega(z)$ is the beam waist radius at the position z , with the beam waist radius at the tip of the fiber ω_0 being 25 μm . z is the distance from the tip of the fiber along the propagation direction. z_R is the Rayleigh range, also known as the Rayleigh length, which is an important parameter for describing the propagation of a Gaussian beam in free space. It is the distance from the beam waist ω_0 (where the beam radius is smallest) to the point where the beam radius increases to $2^{1/2}$:

$$z_R = \frac{\pi \omega_0^2 n_{\text{water}}}{\lambda}, \quad (2)$$

where λ is the wavelength of the incident laser, and n_{water} is the refractive index of water. With these parameters, the intensity distribution of the Gaussian beam exiting the optical fiber in water can be calculated:

$$\begin{aligned} I(r, z) &= \frac{2P}{\pi \omega^2(z)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right) \\ &= \frac{2P}{\pi \omega_0^2 \left[1 + \left(\frac{z}{z_R}\right)^2\right]} \exp\left(-\frac{2r^2}{\omega_0^2 \left[1 + \left(\frac{z}{z_R}\right)^2\right]}\right). \end{aligned} \quad (3)$$

As shown in Fig. 1(d), r is the radial distance from the center of the beam to the position, and P is the beam power. In this study, we have chosen a beam power of 200 mW. The reason for the power selection is explained in section S6 of the SI. Based on the intensity distribution, the spatial distribution of the electric field can be derived.

$$|E(r, z)|^2 = \frac{2I(r, z)}{cn_{\text{water}}\epsilon_0}. \quad (4)$$

$|E(r, z)|^2$ is the square of the electric field intensity at position (r, z) , where c is the speed of light and ϵ_0 is the permittivity of free space.

According to the calculation methods for gradient force and scattering force provided in the SI, the total formula for these forces can be given^{42–45}. The radial gradient force is:

$$F_{\text{grad},r} = \frac{4P\text{Re}(\alpha)r}{cn_{\text{water}}\epsilon_0\pi\omega^4(z)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right). \quad (5)$$

The parameter α in the formula represents the polarizability, the axial gradient force is given by:

$$F_{\text{grad},z} = \frac{4P\text{Re}(\alpha)r^2z(\omega_0^2 - z^2)}{cn_{\text{water}}\epsilon_0\pi\omega^6(z)(\omega_0^2 + z^2)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right), \quad (6)$$

$$\begin{aligned} F_{\text{grad}} &= \left[\left(\frac{4P\text{Re}(\alpha)r}{cn_{\text{water}}\epsilon_0\pi\omega^4(z)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right) \right)^2 \right. \\ &\quad \left. + \left(\frac{4P\text{Re}(\alpha)r^2z(\omega_0^2 - z^2)}{cn_{\text{water}}\epsilon_0\pi\omega^6(z)(\omega_0^2 + z^2)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right) \right)^2 \right]^{1/2}. \end{aligned} \quad (7)$$

The scattering force is:

$$F_{\text{scat}} = \frac{16\pi^2 PR^6}{c\lambda^4 \omega^2(z)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right) \left| \frac{\epsilon_{\text{Ag}} - \epsilon_{\text{water}}}{\epsilon_{\text{Ag}} + 2\epsilon_{\text{water}}} \right|^2, \quad (8)$$

where ϵ_{Ag} represents the dielectric constant of AgNPs, and ϵ_{water} represents the dielectric constant of water. The resultant force on the particle is:

$$F_{\text{total},r} = F_{\text{grad},r} = \frac{4P\text{Re}(\alpha)r}{cn_{\text{water}}\epsilon_0\pi\omega^4(z)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right), \quad (9)$$

$$\begin{aligned} F_{\text{total},z} &= F_{\text{grad},z} + F_{\text{scat}} \\ &= \frac{4P\text{Re}(\alpha)r^2z(\omega_0^2 - z^2)}{cn_{\text{water}}\epsilon_0\pi\omega^6(z)(\omega_0^2 + z^2)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right) \\ &\quad + \frac{16\pi^2 PR^6}{c\lambda^4 \omega^2(z)} \exp\left(-\frac{2r^2}{\omega^2(z)}\right) \left| \frac{\epsilon - \epsilon_m}{\epsilon + 2\epsilon_m} \right|^2. \end{aligned} \quad (10)$$

Results and discussion

Simulation of tapered optical fiber

Through precise optical trapping and manipulation, we can significantly reduce the inter-particle spacing between AgNPs, thereby greatly enhancing the sensitivity and resolution of SERS. To construct a single-beam gradient force optical trap, it is critical to establish a high-gradient optical field. We simulated the intensity distribution of Gaussian beams emitted from variously shaped optical fibers in water using MATLAB. The results revealed that tapered optical fibers, with their gradually narrowing structure and enhanced light field confinement, cause the beam to contract and focus progressively during propagation, achieving a lens-like focusing effect. Detailed simulation results are available in Supplementary information S6. The simulations indicate that the tapered optical fiber is capable of generating a light field with a high convergence angle, largely due to its tapered surface, which allows the beam to achieve a greater convergence angle. This feature concentrates the light field into a small region at the tip of the fiber, where the light intensity reaches its peak. This design enables the precise capture of AgNPs within the high-intensity light field region, significantly reducing the spacing between them. Using the unique properties of the tapered optical fiber, we were able to create a high-gradient optical field, and we employed COMSOL to simulate and analyze its corresponding electric field distribution. Detailed simulation settings and parameters are provided in Supplementary information S7.

Figure 2(a1) and 2(a2) depict the intensity distribution of the 1550 nm laser emitted from the tapered optical fiber in water. The tapered fiber tip was set at 0 μm , and we analyzed the variation in light field intensity along the laser's propagation distance. Figure 2(b1–b8) illustrate the electric field intensity at different positions. The results show that the light field intensity inside the fiber remains relatively constant; however, once the light exits the fiber, the electric field intensity gradually increases, peaking near 2 μm from the tip (as shown in Fig. 2(b4)), before gradually decreasing. This process forms a high-gradient light field at the tip of the fiber, consistent with our theoretical expectations and fulfilling the requirements for creating a three-dimensional optical potential well. Additionally, we observed that the region of enhanced light field intensity forms a spindle-shaped area, which could facilitate the confinement of a large

number of AgNPs within this region.

Simulation of light force on AgNPs

Using the formulas for optical field distribution (Equation (3): $P=200$ mW, $\omega_0=25$ μm), gradient force (Eq. (7)), and scattering force (Eq. (8)), we employed MATLAB to calculate the forces experienced by 50 nm diameter AgNPs within the Gaussian optical field generated by a 1550 nm laser beam emitted from a tapered optical fiber with a diameter of 25 μm . Figure 2(c–f) illustrate the electric field distribution, scattering force, gradient force, and total force acting on these AgNPs. Specifically, Fig. 2(c) shows the electric field distribution of the 1550 nm laser as emitted from the tapered optical fiber in water. The electric field distribution is a critical factor in determining the effectiveness of trapping AgNPs. Figure 2(d) presents the scattering force experienced by 50 nm diameter AgNPs in this optical field. The scattering force results from the interaction of light with the AgNPs, pushing them away from regions of higher light intensity. In Fig. 2(e), the gradient force acting on the AgNPs is displayed. This force, induced by the electric field gradient, directs the AgNPs toward regions of higher field intensity. The magnitude and direction of the gradient force determine the stable position and trapping effectiveness of the AgNPs within the optical field. Figure 2(f) illustrates the total force acting on the AgNPs, which is the vector sum of the scattering and gradient forces. By analyzing the total force distribution, we can deduce the final position and trajectory of the nanoparticles in the optical field. Figure 2(g) demonstrates the strong trapping of 50 nm AgNPs near the tip of the tapered optical fiber. This is because the gradient force, approximately 10^{-18} N, exerted on the AgNPs is significantly greater than the scattering force, approximately 10^{-21} N, as shown in Fig. 2(h). As a result, the total force guides the AgNPs toward the region of highest optical field intensity, located at the tip of the tapered fiber. This substantial reduction in inter-particle spacing significantly enhances the efficiency of SERS detection.

Simulation of AgNPs 'Hot spots'

We developed a COMSOL model to demonstrate how reducing the spacing between AgNPs enhances the sensitivity of SERS. Figure 3(a) presents a simulation of the electric field distribution for 50 nm diameter AgNPs under 532 nm Raman excitation. We analyzed the variations in electric field intensity as the spacing between

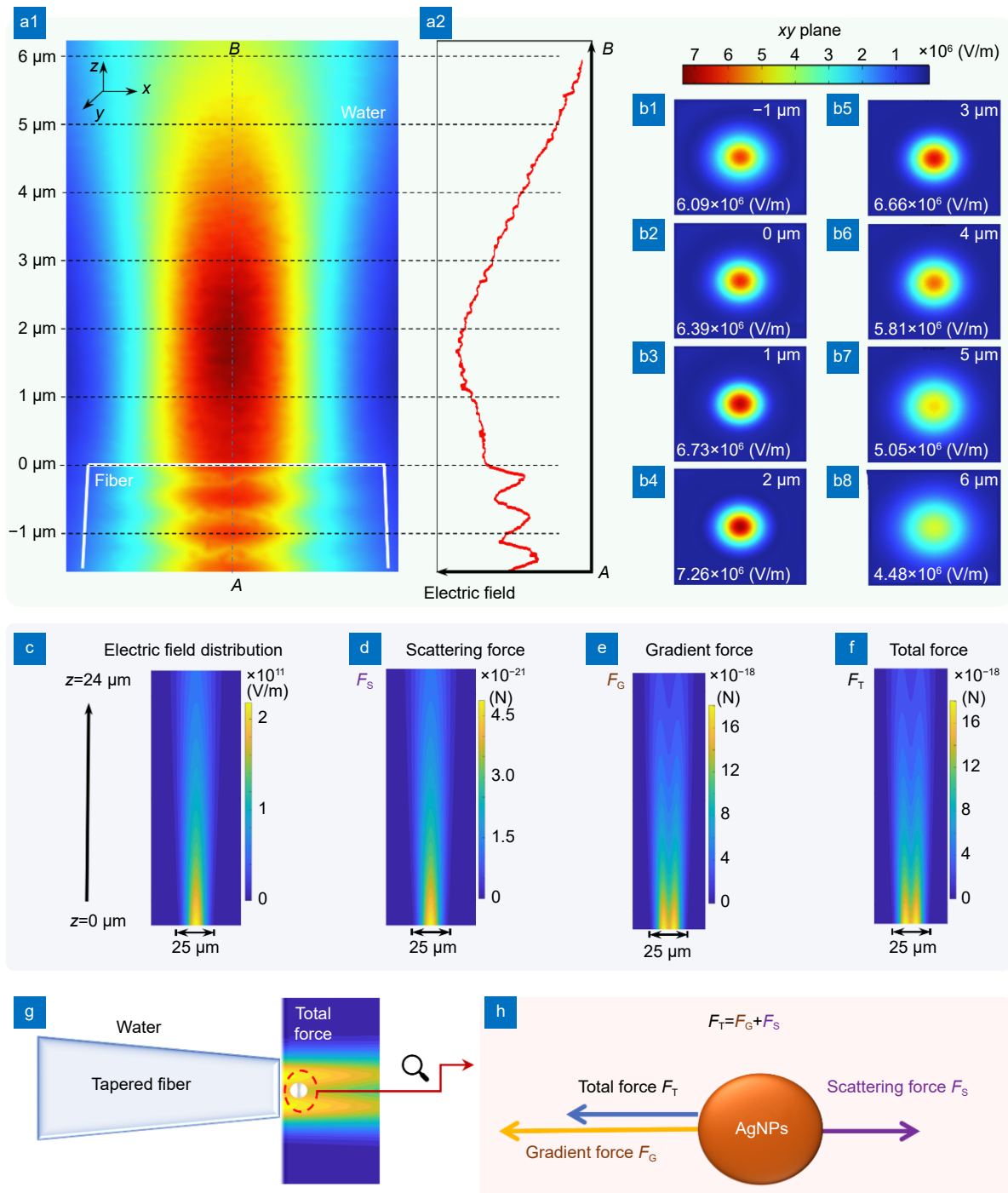


Fig. 2 | (a1, a2) The simulation results of the variation in electric field intensity with distance for the 1550 nm laser emitted from the tapered optical fiber. (b1–b8) Respectively display the electric field intensity distribution at different positions. (c) Electric field distribution of 1550 nm laser emitted by a tapered fiber with a tip diameter of 25 μm in water. (d) Scattering force on 50 nm AgNPs. (e) Gradient force on 50 nm AgNPs. (f) Total force on 50 nm AgNPs. (g) AgNPs are strongly trapped near the tip of the tapered optical fiber. (h) Schematic diagram of the force analysis on AgNPs.

AgNPs decreased. Figure 3(b–g) illustrate these changes under different particle separation conditions. In Fig. 3(b), the AgNPs are trapped near the fiber tip, with a spacing reduced to 1 nm, resulting in a maximum elec-

tric field intensity of 155 V/m. In contrast, Fig. 3(g) shows that when the 1550 nm single-beam optical trap is turned off, the AgNPs become freely dispersed in the aqueous solution, with the spacing increasing to 11 nm

and the maximum electric field intensity dropping to 9.85 V/m. Figure 3(c–f) provide simulation results for intermediate particle gaps of 3 nm, 5 nm, 7 nm, and 9 nm. The corresponding electric field intensities are 59.1 V/m, 38.6 V/m, 14.5 V/m and 12 V/m, respectively. These results clearly indicate that particle spacing plays a crucial role in determining the strength of the electric field. At a 1 nm spacing, the electric field strength peaks at 155 V/m, suggesting strong interactions that enhance the Raman signal. As the spacing increases to 3 nm, 5 nm, 7 nm, 9 nm and 11 nm, the electric field strength decreases to 59.1 V/m, 38.6 V/m, 14.5 V/m, 12 V/m and 9.85 V/m, respectively. This demonstrates that larger separations weaken the interactions between particles, thereby diminishing the enhancement of the Raman signal. Our simulations confirm that activating the single-beam optical trap significantly reduces the interparticle distance between AgNPs, leading to a substantial increase in electric field intensity. These findings indicate that the single-beam optical trap effectively enhances the electric field between AgNPs, improving the detection sensitivity of the optical tweezer-enhanced SERS optofluidic chip.

Raman spectroscopy enhancement capability of the optofluidic chip

Furthermore, we conducted experiments to verify our conclusions. First, we synthesized AgNPs with a diameter of approximately 50 nm using a chemical reduction

method. We then performed Raman molecular fingerprint spectroscopy experiments to validate the enhancement capability of the optofluidic chip for molecular fingerprint detection. In our experiments, we injected the test molecule and prepared AgNPs into the injection port of the optofluidic chip at a 1:1 volume ratio. Two methods were employed for mixing: 1) Real-time mixing method. This method leverages the ability of microfluidic chips to integrate preprocessing steps. By simultaneously injecting the test molecular solution and AgNPs solution at the input port, the turbulent mixing region of the microfluidic chip ensures thorough mixing of the two solutions during flow. This process forms a homogeneous mixture in real time. The online mixing approach significantly enhances reaction efficiency, reduces the steps and time required for preprocessing, and ensures that all components are fully mixed before reaching the optical field region. Consequently, it optimizes signal intensity and improves the uniformity of detection results. 2) Pre-mixing method. In this strategy, test molecule and AgNPs are pre-mixed in a 1:1 volume ratio prior to the experiment to create a uniform mixture, which is then injected into the microfluidic chip. This reduces the mixing time during the experiment, allowing for a quicker attainment of the desired concentration distribution in the optical field. Regardless of the method used, both approaches ensure sufficient contact between crystal violet (CV) and AgNPs. Turbulent mixing occurred in the

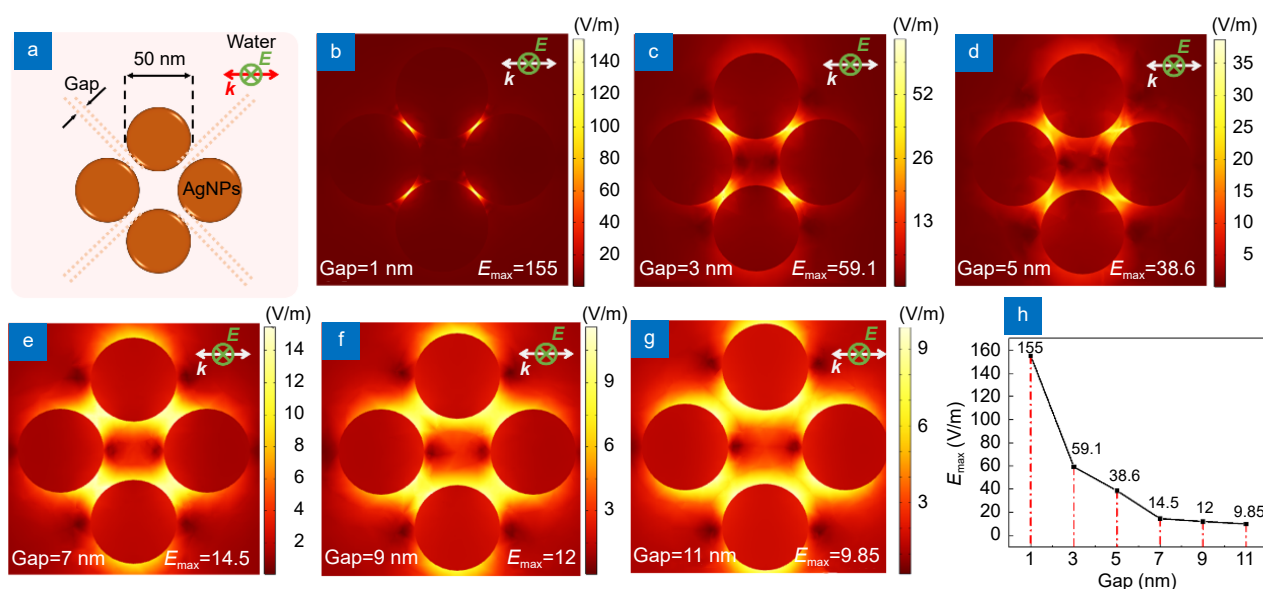


Fig. 3 | (a) Electric field simulation model of 50 nm diameter AgNPs under 532 nm laser excitation with different gaps. (b) Electric field distribution when the gap is 1 nm. Electric field distribution when the gap is (c) 3 nm, (d) 5 nm, (e) 7 nm, (f) 9 nm and (g) 11 nm. (h) The maximal electric field as the gap between AgNPs decreased.

mixing area of the optofluidic chip, forming a mixed solution, which was then injected into the detection area. To observe the trapping effect, we used a confocal Raman microscope to examine how the single-beam optical trap manipulated the mixed solution. Figure 4(a) shows an image captured with the confocal Raman microscope when the optical trap was turned off, revealing a uniformly distributed solution in the detection area. In contrast, Fig. 4(b), captured when the optical trap was turned on, shows that the optical trap emitted from the tapered optical fiber successfully trapped the substances within the mixed solution. Figure 4(c) presents a schematic illustration of how the interparticle distance

between AgNPs decreases when the optical trap is on. Figure 4(d) displays the Raman spectra of 10^{-6} mol/L CV obtained at position 7 as indicated in Fig. 4(b), comparing the spectra when the optical trap was turned on and off. Figure 4(e) highlights the intensity of Raman characteristic peaks at 913, 1177, and 1621 cm^{-1} from Fig. 4(d). When the optical trap was off, the intensities of these peaks were 1517 a.u., 1178 a.u., and 1653 a.u., respectively. However, when the optical trap was activated, the intensities of these peaks increased significantly to 2910 a.u., 2127 a.u., and 3176 a.u., nearly doubling the signal strength. These results demonstrate the effectiveness of the optical trapping system. By controlling the on-off

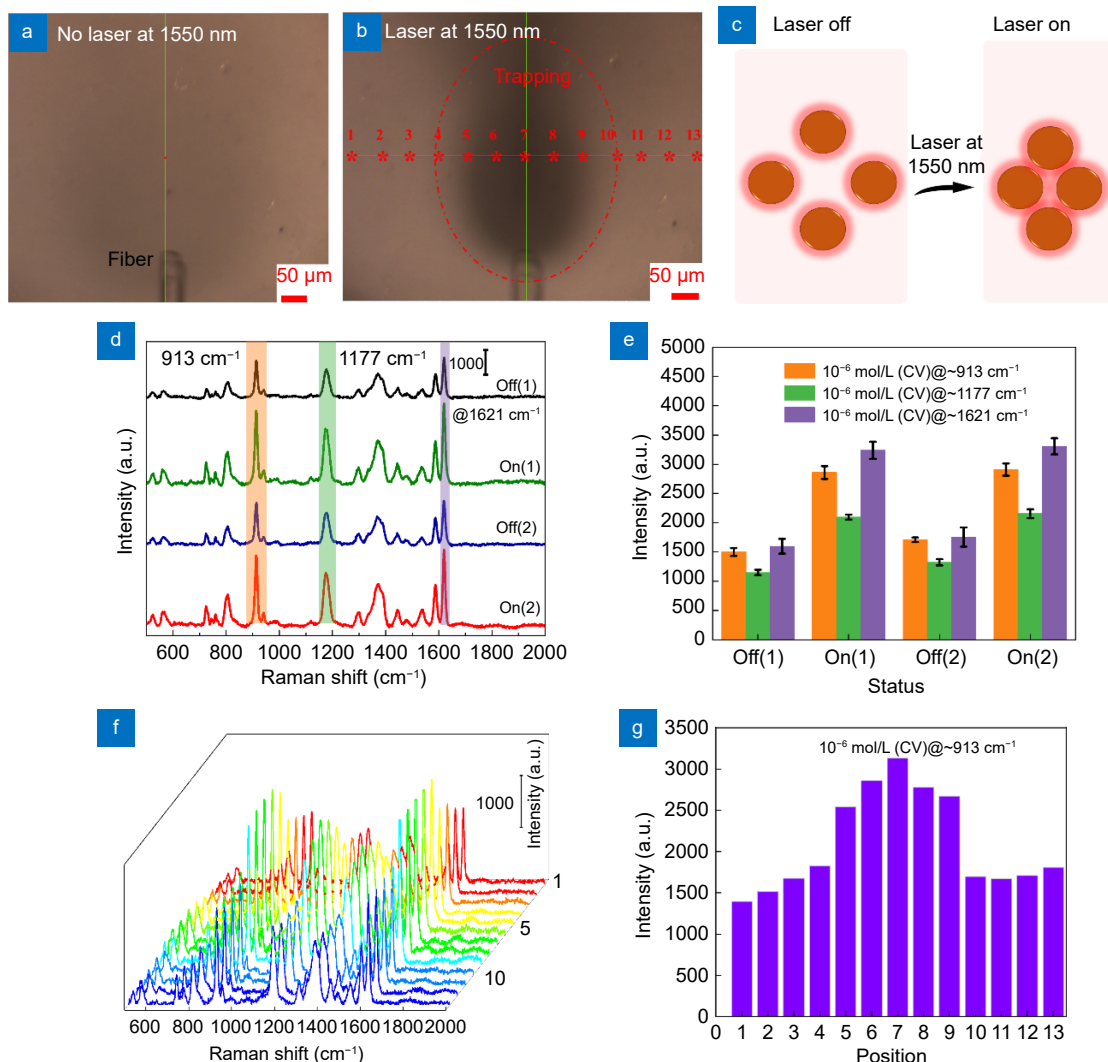


Fig. 4 | Image observed by confocal Raman spectrometer microscope (a) when the single-beam optical trap is closed, with the detection region filled with a homogeneous and stable mixed solution. (b) When the single-beam optical trap is opened, with substances in the mixed solution captured by the optical trap emitted from the tapered fiber optical fiber. (c) Schematic diagram of AgNPs gap when the single-beam optical trap is off and on. (d) Raman spectra of 10^{-6} mol/L CV obtained by opening and closing the single-beam optical trap of position 7 in (b). (e) Raman characteristic peak intensity at 913, 1177 and 1621 cm^{-1} in (d). (f) Raman line mapping obtained at different positions when the optical trap is open. (g) Raman characteristic peak intensity at 913 cm^{-1} in (f).

state of the single-beam optical trap, we could manipulate particle trapping and observe significant differences in Raman signal intensity, confirming the enhancement provided by the optical trap in SERS detection.

We further utilized Raman line mapping function to analyze the Raman molecular fingerprint spectra in non-laser trapping areas, in laser trapping areas, and in regions that back to non-laser trapping. Each detection point was spaced 50 μm apart. Figure 4(b) clearly shows the relative layout of each detection point and the laser trapping area. Closer examination of the Raman molecular fingerprint spectra of CV at each point in Fig. 4(f) reveals that the Raman signal intensity in the laser trapping area is significantly higher than in the non-laser trapping area. We present the comparative results of the effect of AgNPs spacing on electric field intensity and Raman signal enhancement in section S8 of the supporting information. To more intuitively observe the intensity changes of the Raman characteristic peaks, we specifically focused on the characteristic peak at 913 cm^{-1} and found that its intensity varied significantly, increasing as the distance to the center of the laser trapping area decreased (Fig. 4(g)). Based on this finding, we conducted further Raman mapping experiments.

In Fig. 5(a), we marked the detection locations for the experiment and Fig. 5(b) shows the intensity of the Raman characteristic peak at 913 cm^{-1} at different positions marked in Fig. 5(a). It is evident that the Raman molecular fingerprint spectrum intensity in the laser trapping area is much higher than in the non-laser trapping area. Notably, the laser trapping area shown in Fig. 5(a) is larger than that in Fig. 4(b). There are two main reasons for this: (1) The incident beam diameter of the 532 nm Raman spectrometer we used is only 1 micrometer, which may have affected the stable trapping of the single-beam optical trap during mapping, promoting thermophoretic motion of the AgNPs. (2) Since the Raman mapping test took a relatively long time, the concentration gradient caused more particles to be trapped in this area. Finally, Fig. 5(c) and Fig. 5(d) illuminate the detection limit of the chip. By utilizing the laser trapping technique, we successfully detected the Raman spectrum of CV at a concentration of 10^{-9} mol/L (Fig. 5(c)), demonstrating the effectiveness of the single-beam optical trap in enhancing the molecular fingerprint spectrum identification capability of the SERS optofluidic chips. Additionally, we expanded the experimental scope to include the pesticide thiram, successfully detecting

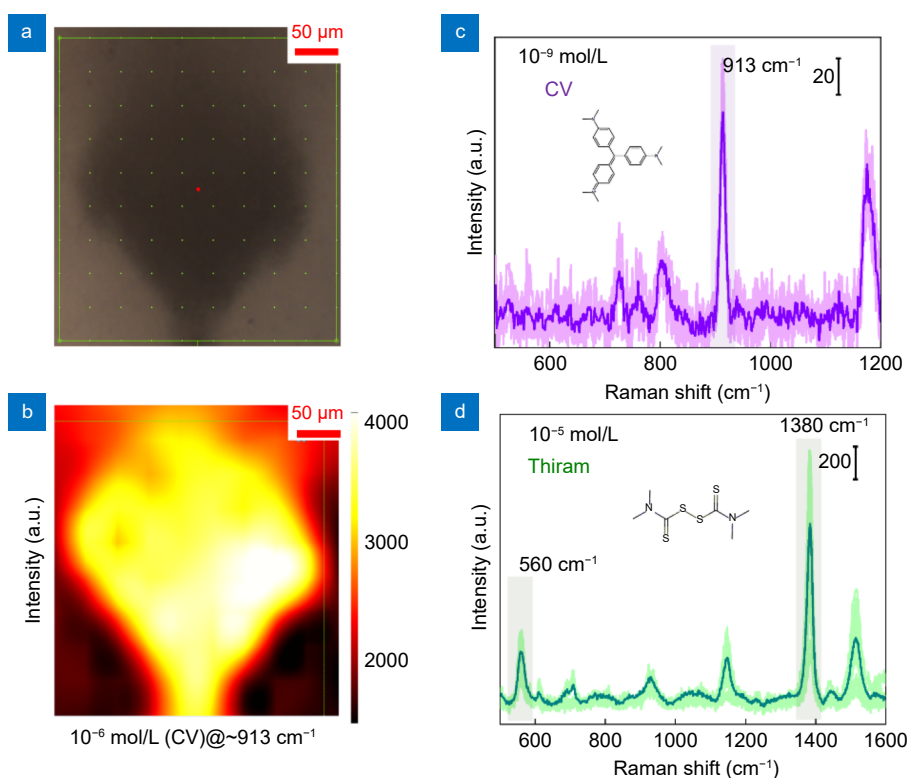


Fig. 5 | (a) Positions of the Raman mapping. (b) Raman characteristic peak intensity at 913 cm^{-1} at different positions in (a). (c) Detection limit of the system for CV at a concentration of 10^{-9} mol/L. (d) Detection limit of the system for thiram at a concentration of 10^{-5} mol/L.

concentrations as low as 10^{-5} mol/L (Fig. 5(d)). These findings highlight the feasibility of our system for routine water quality monitoring, particularly in detecting pesticide residues, showcasing its significant potential for environmental applications.

Conclusions

In this study, we successfully developed a photonics-enhanced SERS optofluidic detection system. Leveraging the high mixing efficiency and high-throughput characteristics of the optical microfluidic chip, the system enables thorough mixing of small sample volumes with Ag-NPs, significantly reducing sample consumption. Based on this, the enhancement of SERS signals is concentrated within the optical trap region, substantially improving the detection signal intensity. Furthermore, by integrating the tapered optical fiber into the optical microfluidic chip, the system achieves stable fixation of the optical trap region, greatly simplifying the experimental workflow. Users only need to inject the analyte and Ag-NPs into the chip and activate the optical trap laser to perform high-sensitivity detection at the fiber tip region. Through comprehensive analysis, we employed COMSOL simulation software to model the optical field distribution in the tapered optical fiber, and established a MATLAB-based theoretical framework to validate the effectiveness of the single-beam optical trap in capturing AgNPs. These simulations confirmed the theoretical feasibility of our model. Experimentally, we controlled the on-off states of the optical trap to investigate particle capture and release dynamics. The results demonstrated that Raman signal intensity in the capture state was significantly higher than in the non-capture state. This observation verified that the single-beam optical trap effectively enhances the detection capabilities of the SERS optofluidic system. To further examine the influence of the trapping area on the SERS effect, we employed Raman mapping techniques. These results indicated a substantial enhancement in the SERS signal when optical trapping was applied, confirming the effectiveness of the optical trap in improving the system's detection sensitivity. Notably, the optical trapping-enhanced SERS optofluidic detection system developed in this study has the potential to be integrated with portable high-power lasers and advanced Raman spectrometers. This integration is pivotal for developing highly compact, efficient, and portable optoelectronic sensing technologies, broadening the application range in fields such as biomedical

diagnostics, environmental monitoring, and food safety. The advancements in portability and performance of this optoelectronic system are essential for facilitating real-world applications of SERS technology.

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Author contributions

N. S. and J. Z. designed the study, discussed the study results, contributed to the interpretation of the data. N. Sun conducted experiments and wrote the paper. Y.G, Y.W, X.W, Z.Y. and S.S. provided support in experiments.

Competing interests

The authors declare no competing financial interests.

Supplementary information

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