

Supplementary Information S1: Detailed explanation of the EF calculation.

We describe how to calculate the enhancement factor (EF).

$$EF = \frac{I_{SERS}/N_{SERS}}{I_{BULK}/N_{BULK}}$$

$I_{SERS}$  = 7415 for the silk fiber and 9337 for the silica fiber

$I_{BULK}$  = 10,095 for the 4-ABT power

$$N_{SERS} = \frac{\text{Total surface area of gold nanoparticles in laser spot size}}{\text{Size of target molecule}} = 35.55 \times 10^9$$

$$N_{BULK} = \frac{D_{molecule} \times \text{Laser spot size} \times \text{Penetration depth}}{MW_{molecule}} \times 6.022 \times 10^{23} = 89.16 \times 10^{13}$$

Table S1. Calculation parameters

Diameter of laser	100 $\mu\text{m}$
Penetration depth	20 $\mu\text{m}$
Density of 4-ABT ( $D_{molecule}$ )	1.18 g/cm <sup>3</sup>
Molecular weight of 4-ABT ( $MW_{molecule}$ )	125.19 g/mol
Size of 4-ABT	0.2 nm <sup>2</sup>
Total surface area of gold nanoparticles	7.11 m <sup>2</sup>

$$EF (\text{silk fiber}) = \frac{I_{SERS}}{N_{SERS}} \times \frac{N_{BULK}}{I_{BULK}} = 1.84 \times 10^4$$

$$EF (\text{silica fiber}) = \frac{I_{SERS}}{N_{SERS}} \times \frac{N_{BULK}}{I_{BULK}} = 2.31 \times 10^4$$