Supporting information for

Leaky Mode Resonance of Polyimide Waveguide Couples Metal Plasmon Resonance for SERS

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1. Preparation of PI films with different thickness.

To accurately control the thickness of the PI film, we changed the concentration of the PI solution (75% and 100%) and keep the spinning speed at a certain rate. Tables S1 and S2 list the thicknesses of PI films with multi layers measured by a probe profiler (Veeco, Dektak 150).

Table S1 The thickness of PI with multi layers of the PI solution of 75%.

Times of spinning	Value ± Error (nm)
Once	230 ± 4
Twice	250 ± 7
Three times	260 ± 9
Four times	270 ± 5

Table S2 The thickness of PI with multi layers of the PI solution of 100%.

Times of spinning	Value ± Error (nm)
Once	290 ± 4
Twice	450 ± 8
Three times	580 ± 7
Four times	690 ± 9

2. Measurement of PI refractive index and thickness by ellipsometry.

The original data were obtained by a ellipsometer (Uvisel Ⅱ, Horiba). By fitting the data through the settings "Range: 1.5000-6.0000 eV, Increment: 0.0500 eV, Points Number: 91", a five layers structure was established along with the process searching, including air, PI, PI/Ag interface, Ag and glass layers as shown in Scheme S1, in which the PI layer was described by a dispersion formula of New Amorphous (short for "nam") derived by Horiba Jobin Yvon from Forouhi-Bloomer formulation.
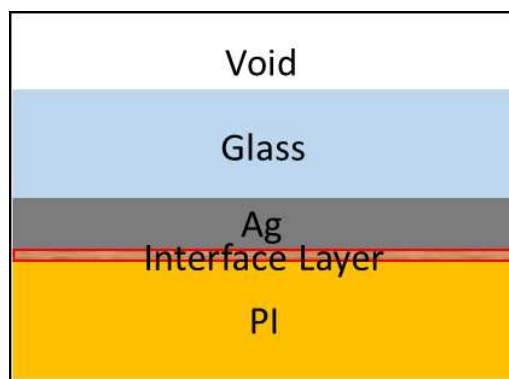
Scheme S1. The optical model for ellipsometric data fitting.

The fitting and result information shown in the following panel is a PI film obtained by spinning coating PI (100%) at a speed of 700 rpm for 10 s and 2000 rpm for 20 s. The resulted Root-Mean-Square of 10.02 within the range of 1.5-5.5 eV exhibits accessible correspondence between measured and fitted data. Additionally, the absolute value of every single number in correlation matrix less than or equal to 0.835 means an excellent independence between every two parameters. The resulted fitting dispersive curve gave an exact value of 1.760 at the wavelength of 532 nm.

Table S3 Parameters of simulated waveguide

Layers	Value±Error (nm)
Ag film thickness	27.738 ±0.986
Interface layer thickness	11.327 ± 1.382
PI layer thickness	278.656 ± 3.310

Table S4 Correlation matrix calculated between the parameters used for fit.

=1=	=2=	=3=	=4=	=5=	=6=	=7=	=8=
1	-0.056	-0.382	0.218	-0.471	-0.228	-0.039	0.037
	1	0.2228	-0.547	-0.364	0.023	0.321	0.282
		1	-0.821	-0.267	-0.274	0.500	0.104
			1	0.638	0.337	-0.835	-0.239
				1	0.585	-0.771	-0.127
					1	-0.507	0.694
						1	0.093
							1

Eight parameters listed are Ag film thickness, Interface layer thickness, PI layer thickness, $n\infty$, ω_j, f_j, ω_g and Γ_j in nam dispersion formula of PI.
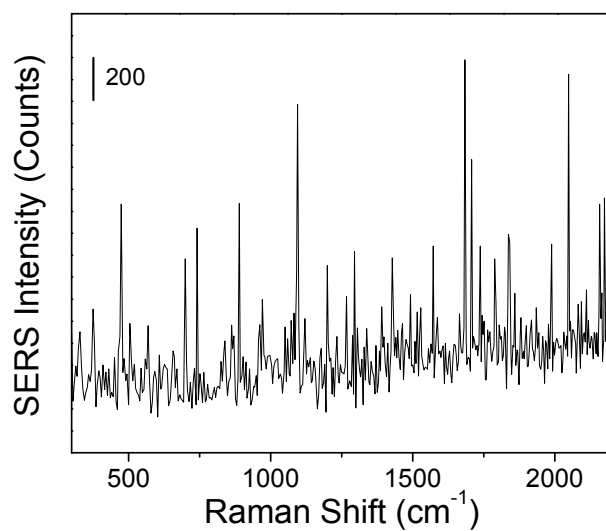
3. Raman spectrum of PI.

Figure S1 Raman spectrum of PI under the excitation wavelength of 532 nm. The power of the laser was 1.5 mW and the integration time of the CCD was 15 s.

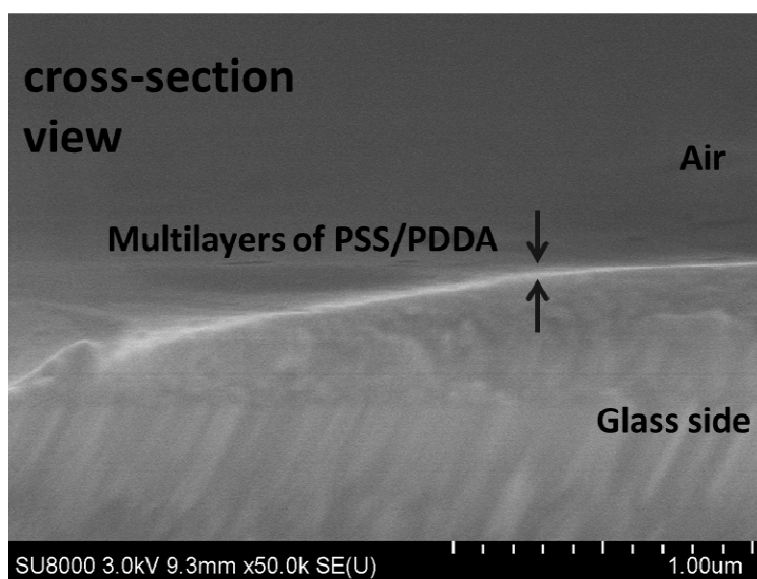**4. The measure of the thickness of a bilayer of PSS/PDDA**

Figure S2 The SEM image of ten bilayers of PSS/PDDA. The total thickness of multilayers is about 21 nm.

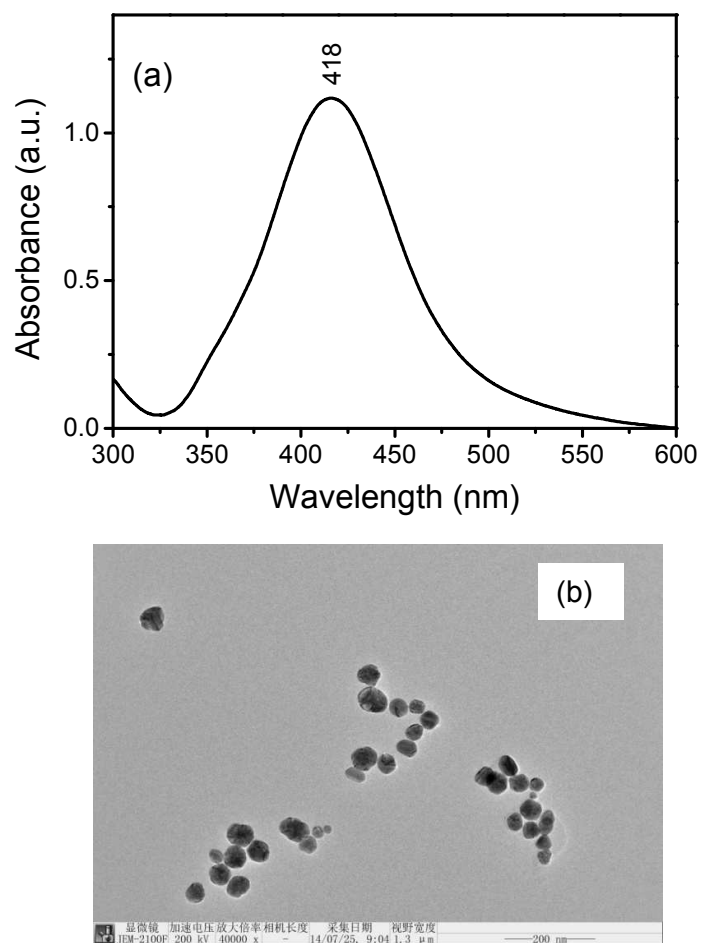## 5. Uv-vis spectrum and TEM image of Ag nanoparticles.

Figure S3 Uv-vis spectrum (a) and the TEM image (b) of the prepared Ag colloid.

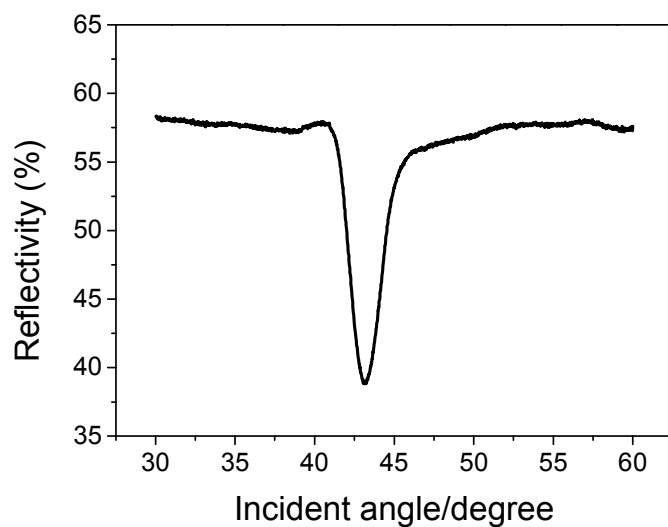**6. The resonance curves of the prism-type SPR excitation at 532 nm.**

Figure S4 The resonance curves of the prism-type SPR excitation at 532 nm. The thickness of Ag film is 40 nm and the resonance angle is 43.2°.

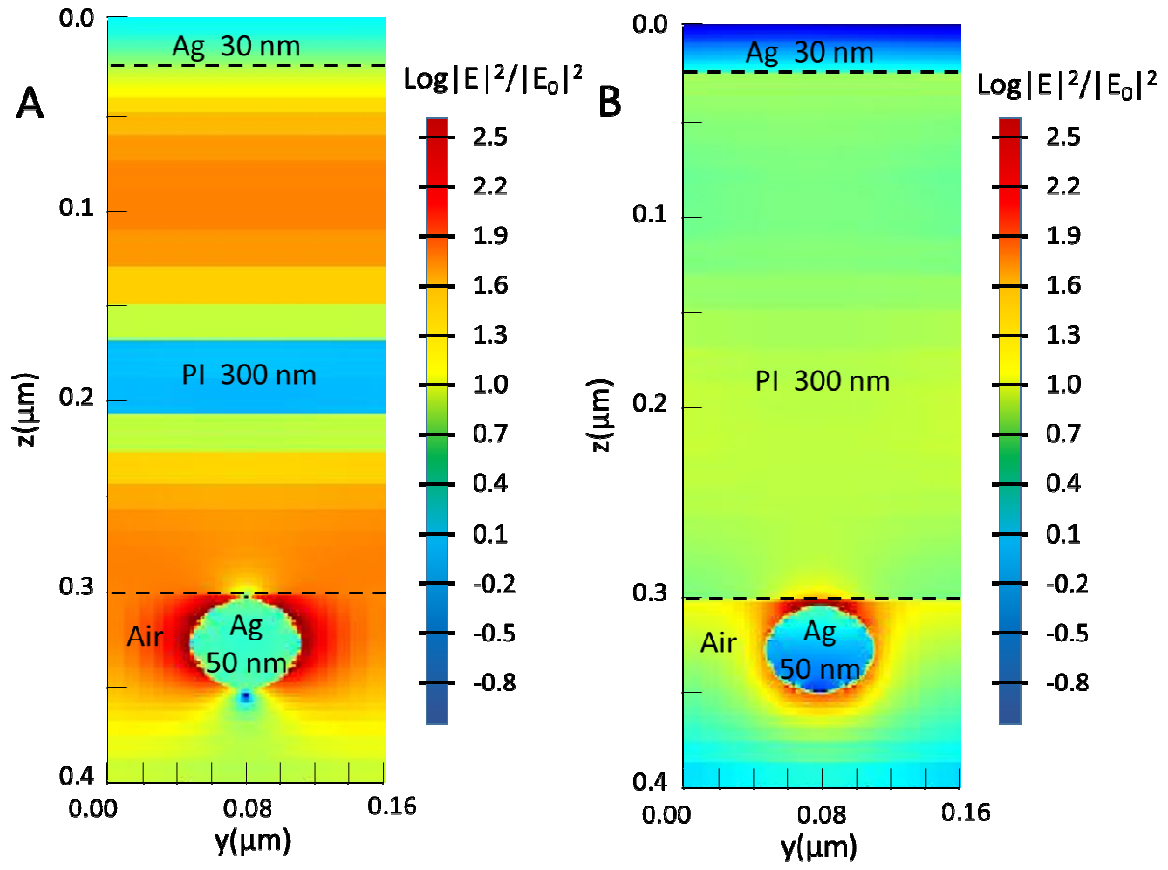**7.** Comparison of the excitation modes of TE and TM.

Figure S5 FDTD simulated EM field distributions of an Ag nanoparticle assembled on the PI waveguide (thickness =300 nm) surface in TE1 (A) and TM1 (B) mode with the 532 nm light source. The incident angles for TE1 and TM1 are 49.6° and 41.9°, respectively.
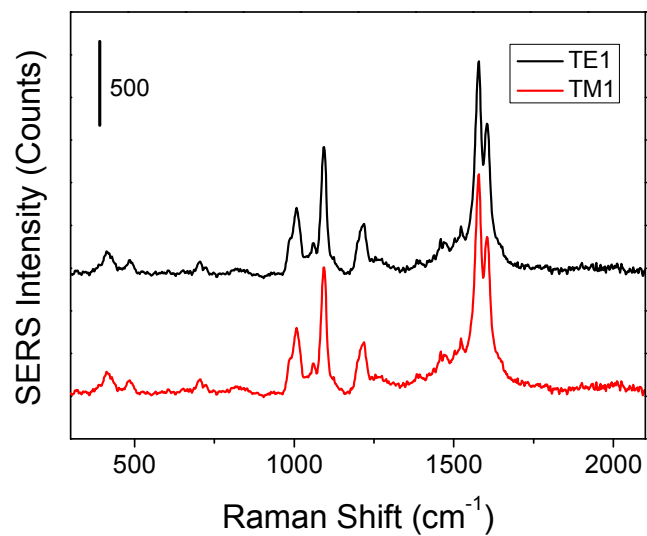
Figure S6 SERS spectra of 4-Mpy (1.0×10^{-6} M) under the LMR-MPR coupling containing in TE1 mode (black line) and in TM1 mode (red line). The power of the laser was 1.5 mW and the integration time of the CCD was 15 s. The incidence angles for TM1 and TE1 are 43.5° and 50.4°, respectively.