

## **Controlled Construction of Nanodisk Dimer-over-Mirror for Near-Field Enhancement and Linewidth Shrinking**

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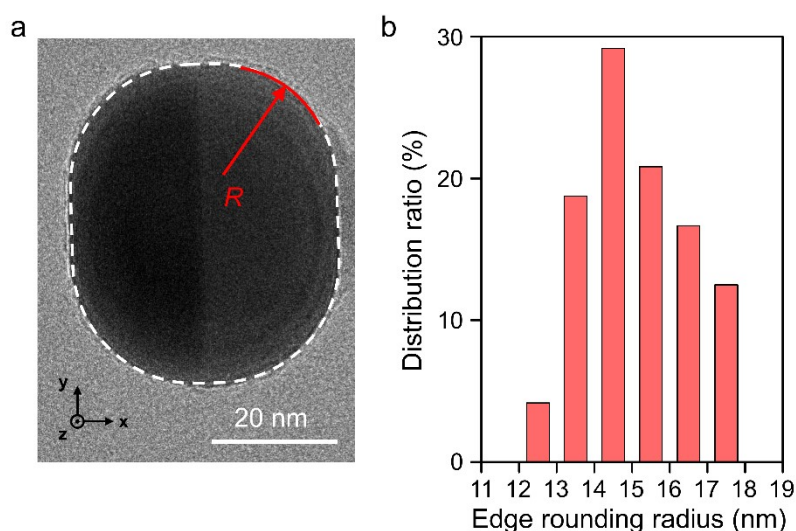
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## 1. Edge rounding curvature of nanodisk

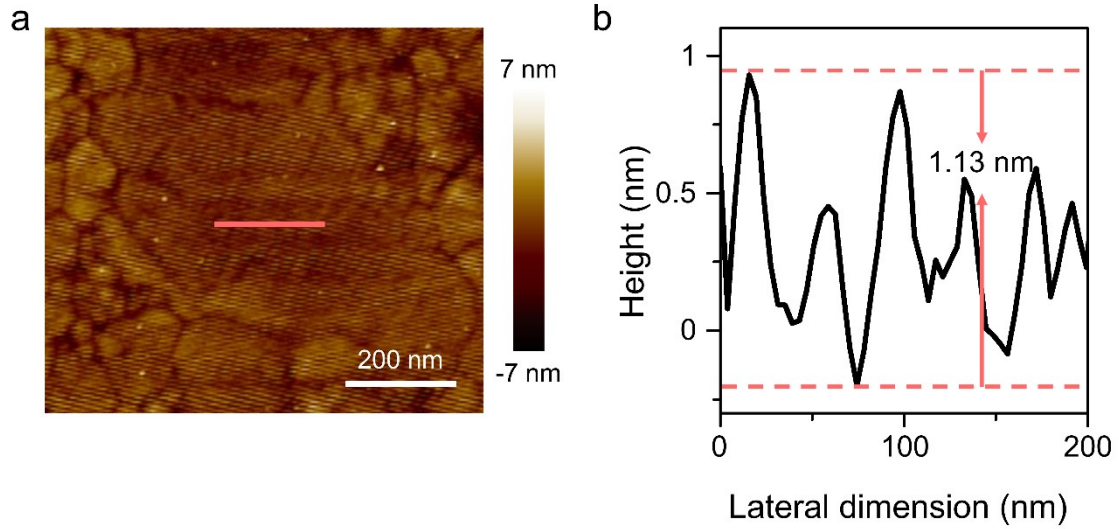
The side wall of the vertically oriented gold nanodisk was characterized by the transmission electron microscopy, with an average edge rounding radius of 15.0 nm with a deviation of 1.5 nm (**Fig. S1**).



**Fig. S1** (a) Transmission electron microscopy image of the gold nanodisk. (b) Statistical histogram about the edge rounding radius.

## 2. Surface roughness of Al<sub>2</sub>O<sub>3</sub>-coated silver film

The surface roughness of the 1 nm-Al<sub>2</sub>O<sub>3</sub>-coated silver film was measured with an atomic force microscope. The root-mean-square roughness was 0.54 nm over an area of 100 × 50 nm<sup>2</sup> covered by a nanodisk dimer (**Fig. S2**). The atomic force microscope image further revealed that the typical height difference within the relatively flat regions reached 1.13 nm.



**Fig. S2** (a) Atomic force microscope image of 1 nm- $\text{Al}_2\text{O}_3$ -coated silver film. (b) Height profiles of the underlined part in (a).

### 3. Comparison of quality factors for different plasmonic nanocavities

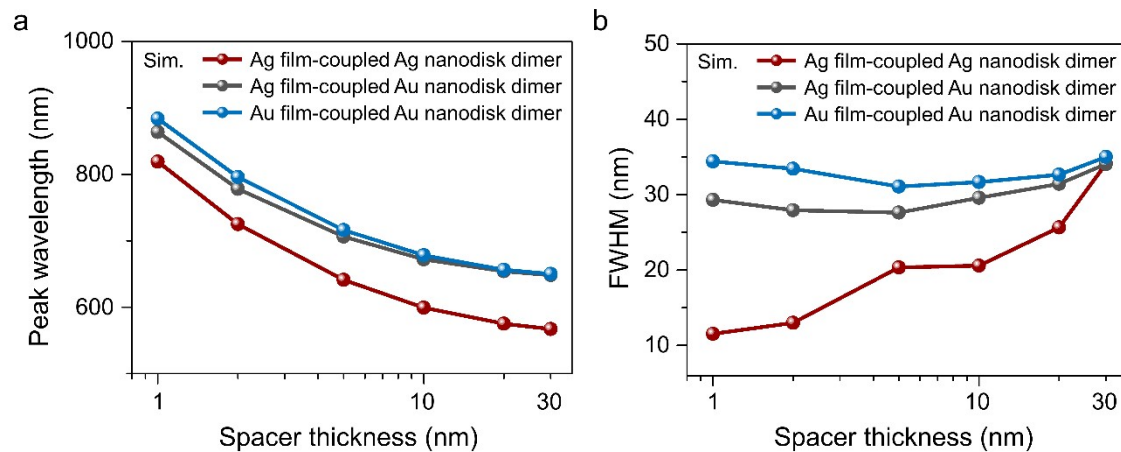
**Table 1** Comparison of the quality factors for various plasmonic nanocavity structures

Plasmonic nanocavity	Quality factor	Reference
Gold nanosphere	7	Phys. Rev. Lett. 88, 077402 (2002)
Gold nanosphere dimer	8	Nat. Biotechnol. 23, 741 (2005)
Gold nanosphere over gold film	9	Nature 535, 127 (2016)
Gold nanosphere dimer over gold film	19	Laser Photonics Rev. 14, 2000068 (2020)
Silver nanorod (third-order mode)	29	Nanoscale 5, 6985 (2013)
Silver nanocube over silver film	15	Nano Lett. 14, 4797 (2014)
Gold nanodisk dimer	4.4	Phys. Rev. Lett. 124, 163901 (2020)
Film-coupled nanodisk dimer	18.4	This work

### 4. Comparison of three different film-coupled nanodisk dimer systems

Simulations of three configurations ( $D = 50$  nm) reveal the M-mode peak wavelength and full width at half maximum (FWHM) versus spacer thickness ( $t = 1 - 30$  nm). As spacer thickness increases from 1 nm to 30 nm, the Ag film-coupled Ag

nanodisk dimer shows a peak wavelength shift from 818 nm to 566 nm with a FWHM range of 10.4 – 33.0 nm; the Ag film–coupled Au nanodisk dimer exhibits a peak shift from 864 nm to 648 nm with a FWHM range of 27.6 – 34.0 nm; the Au film–coupled Au nanodisk dimer displays a peak shift from 884 nm to 650 nm with a FWHM range of 30.9 – 34.9 nm.



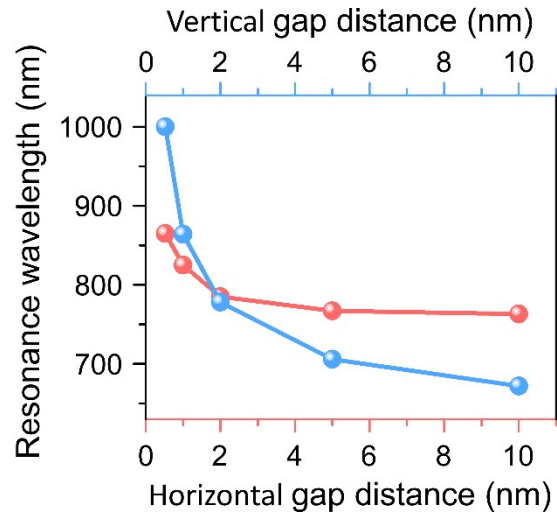
**Fig. S3** Simulated peak wavelength (a) and FWHM (b) of the M mode as a function of spacer thickness for three different systems: Ag film–coupled Ag nanodisk dimer (red), Ag film–coupled Au nanodisk dimer (grey), and Au film–coupled Au nanodisk dimer (blue).

## 5. Dependence of resonance wavelength on gap geometry

Based on classical electromagnetic theory, the M-mode resonance wavelength of a 50-nm-diameter film-coupled nanodisk dimer was simulated to investigate the dependence of the resonance wavelength on the horizontal and vertical gap distance. As shown in **Fig. S4**, with the horizontal gap fixed at 0.52 nm, widening the vertical gap from 0.52 nm to 10 nm shifts the resonance wavelength from 1000 nm to

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672 nm; conversely, with the vertical gap fixed at 1 nm, extending the horizontal gap over the same range shifts it from 864 nm to 762 nm.



**Fig. S4** Comparison of M-mode resonance wavelengths for varying horizontal (red) and vertical (blue) gap distances.