Supporting Information for

Hybridization of Epsilon-Near-Zero Modes via Resonant Tunneling in Layered Metal/Insulator Double Nanocavities

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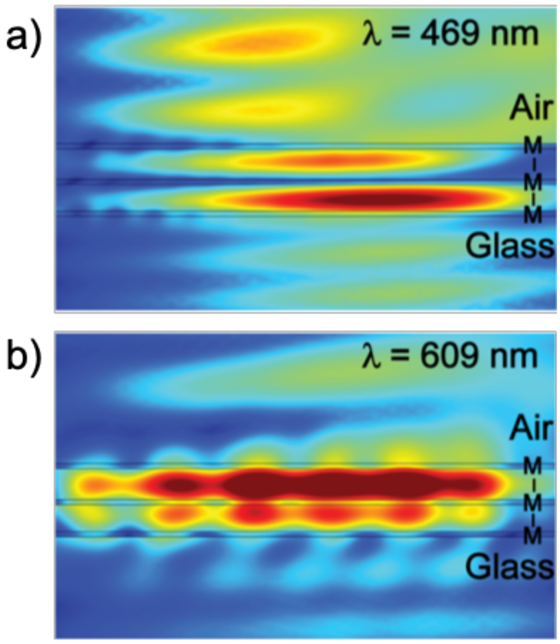

Figure S1. Normalized amplitude of the electric field in the MIMIM nanocavity (with geometrical parameters as in Fig.1) for incident light at an angle of θ=40°, calculated by Finite Element Method (COMSOL) simulations at the two ENZ wavelengths.

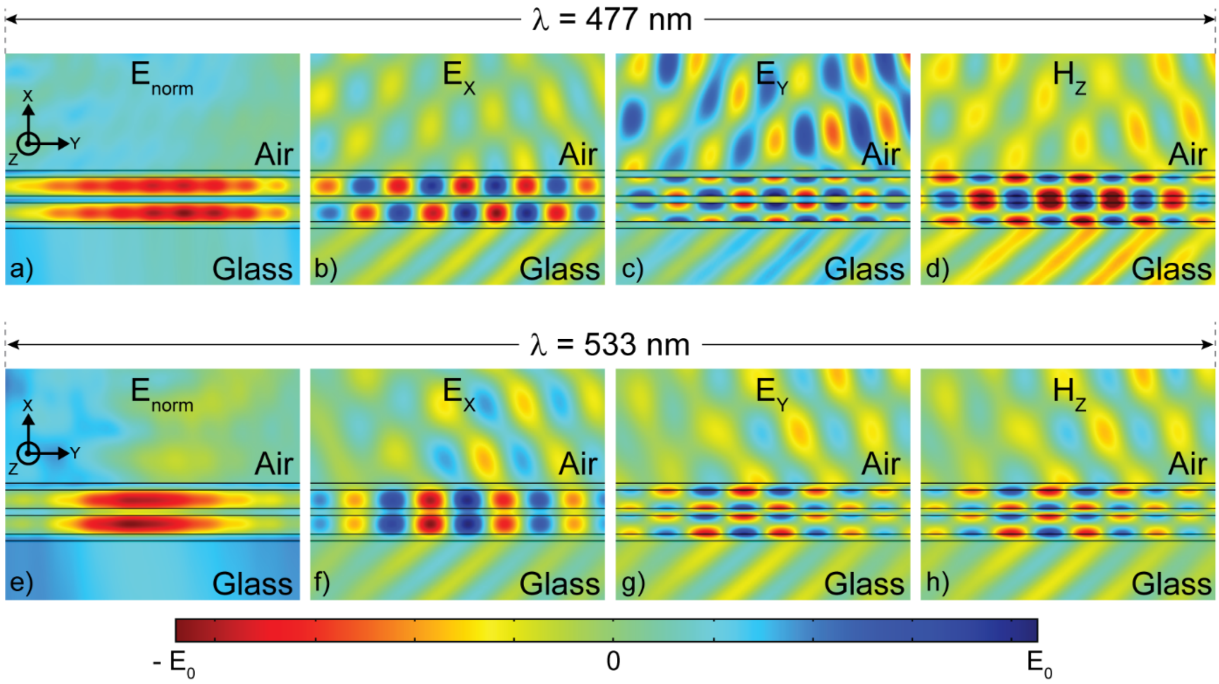

Fig. S2: COMSOL calculated cross sections of the MIMIM structure (with 40 nm metal layer thickness, and 110 nm thick dielectric layers) showing the (a,e) norm, (b,f) X-component, (c,g) and Y-component of the electric field for the odd (a-d) and even (e-h) modes. (d,h) Z-component of the magnetic field for the odd (d) and even (h) modes.

Figures S2 (a,e) show the electric field norm for the odd (a) and even (e) cavity modes, demonstrating the field confinement inside the cavity at the two resonant wavelengths. The x-component of the E field of the odd mode (b) holds exactly the features of the odd mode of a double potential quantum well and the same can be said for the even mode (f). These two modes can be seen also as the out-of-phase (odd) or in-phase (even) superposition of the resonances of two coupled MIM cavities. The y component of the electric field, as well as the z component of the magnetic field show the longitudinal character of the waveguided mode, highlighting its similarity with the Ferrell-Berreman modes.

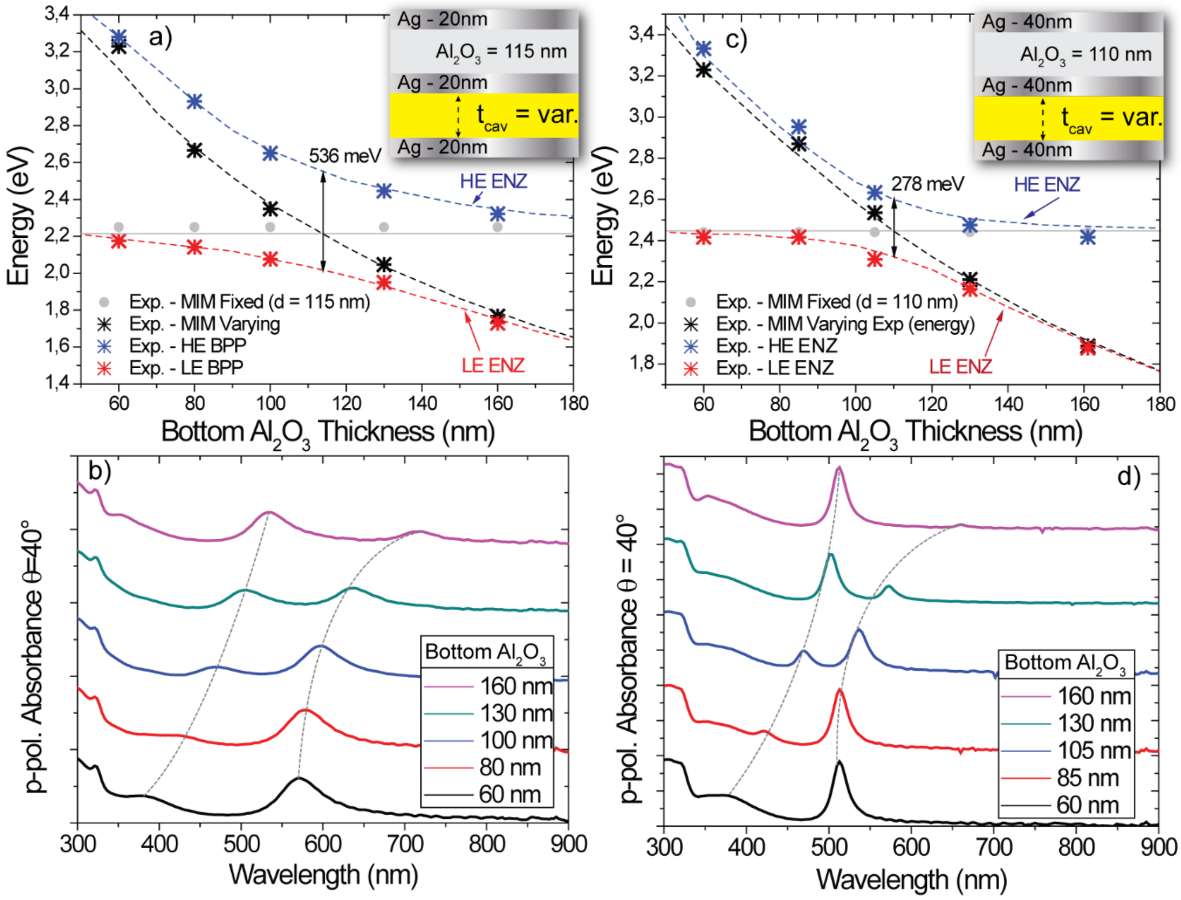

Figure S3. Mode anticrossing in the experimentally measured absorbance spectra for MIMIM cavities with Ag layers of 20 nm (a,b) and 40 nm thickness (c,d), resulting in a mode splitting of (a,b) 536 meV, and of 278 meV for Ag layers, respectively.

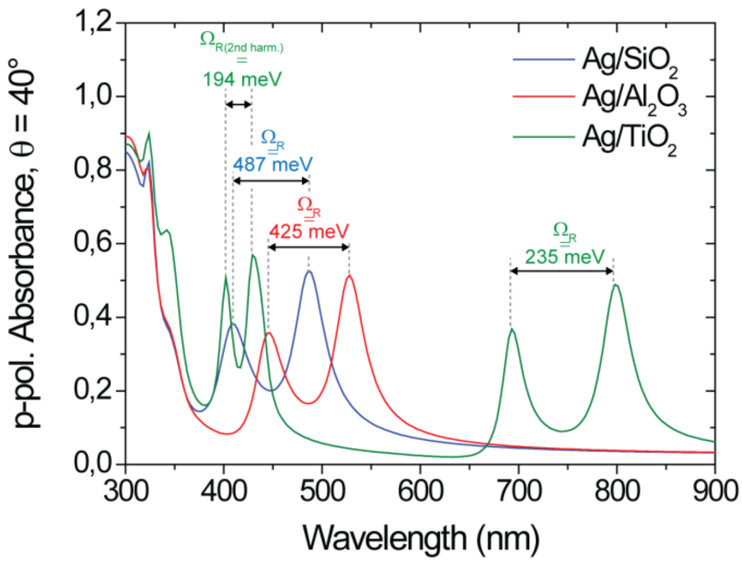

Figure S4. SMM simulated absorbance spectra of MIMIM cavities with SiO_2=100nm (blue curve), Al_2O_3=100nm (red curve) and TiO_2=100nm (green curve) as dielectric material, and thickness of Ag layers of 30nm. Interestingly, the very high refractive index of TiO_2 allows for the occurrence of second order harmonic resonances, revealing that also the higher order modes manifest a mode splitting. An increase in the refractive index of the dielectric layers leads to a red-shift of the ENZ resonances.

Exact analytic expression for the bound and quasi-bound modes of the double-potential-well constituted by the MIMIM

Due to the symmetry of the potential, the "parity" operator commutates with the "potential energy" operator, so that symmetric eigenmodes can be found by solving the Schrödinger Equation in each domain matching the solutions and their derivatives at the boundaries. An educated guess of the shape of the even and odd solutions can be done as shown in Figure 4b. The wavefunction in each region can be expressed as follows:

Region I (central metal – separation barrier):

The symmetric solution of the Schrödinger Equation in this region is well modeled by a hyperbolic cosine function:

$$\psi(x) = \cosh(k_0 \kappa x), \qquad \text{S2}$$

Region II (dielectric layer - well):

The symmetric solution of the Schrödinger Equation is a sinusoidal function with an additional phase component:

$$\psi(x) = \sin(k_0 n x + \phi), \qquad \text{S3}$$

Region III (external metal barrier):

In the thick metal, the wavefunction decays exponentially, therefore:

$$\psi(x) = C\exp(k_0 \kappa x), \qquad \text{S4}$$

The implicit dispersion relation can be found by solving the system in Eq. S5a-d:

$$\begin{cases} A\cosh(k_0\kappa \frac{t_m}{2}) = B\sin(k_0 n \frac{t_m}{2} + \phi), & \text{(S5.a)} \\ A\,k_0\kappa\sinh(k_0\kappa \frac{t_m}{2}) = B\,k_0 n\cos(k_0 n \frac{t_m}{2} + \phi), & \text{(S5.b)} \\ B\sin(k_0 n\, t_{TOT} + \phi) = C\exp(-k_0\kappa t_{TOT}), & \text{(S5.c)} \\ B\,k_0 n\cos(k_0 n\, t_{TOT} + \phi) = -C\,k_0\kappa\exp(-k_0\kappa t_{TOT}), & \text{(S5.d)} \end{cases}$$

Where $t_{TOT}=t_m/2+t_d$, with t_d as the thickness of the dielectric layer, and t_m the one of the metal layer. Dividing Eq. S5.a by Eq. S5.b, and Eq. S5.c by Eq. S5.d we obtain:

$$\begin{cases} \frac{1}{\kappa}\tanh(k_0\kappa \frac{t_m}{2}) = -\frac{1}{n}\tan(k_0 n \frac{t_m}{2} + \phi), & \text{(S5.e)} \\ \tan(k_0 n\, t_{TOT} + \phi) = -\frac{n}{\kappa}, & \text{(S5.f)} \end{cases}$$

It is possible to calculate the value of the phase component simply by expliciting Eq. S5.f:

$$k_0 n\, t_{TOT} + \phi = \arctan\left(-\frac{n}{\kappa}\right), \qquad \text{(S5.g)}$$

For small angles, the arctangent function can be approximated with its argument, so that:

$$k_0 n\, t_{TOT} + \phi \sim -\frac{n}{\kappa}, \qquad \text{(S5.h)}$$

And, finally:

$$\phi \sim -n\left(\frac{1}{\kappa} + k_0 t_{TOT}\right) = \left(-\frac{n}{\kappa} - nk_0\frac{t_m}{2} - k_0 n t_d\right), \qquad \text{(S5.i)}$$

Replacing Eq. S5.i in Eq. S5.e,f

$$\tanh(k_0\kappa \frac{t_m}{2}) = \frac{\kappa}{n} \tan\left[-k_0 n \left(\frac{1}{k_0\kappa} + t_d \right) \right], \qquad \text{(S6)}$$

Straightforwardly, the implicit dispersion relation for the antisymmetric modes comes by replacing the hyperbolic cosine with a hyperbolic sine function and, therefore:

$$-\coth(k_0\kappa \frac{t_m}{2}) = \frac{\kappa}{n} \tan\left[-k_0 n \left(\frac{1}{k_0\kappa} + t_d \right) \right], \qquad \text{(S7)}$$

Equation S6 and S7 represent the analytic dispersion for the thick walls double potential well (MIMIM cavity).

When the thickness of the external metals reduced below 40 nm, the tunneling probability through them is no more negligible, and an additional phase component has to be considered that accounts for the finite tunneling probability. Therefore, the solutions in *Region II* are equal to:

Region II (dielectric layer - well):

$$\Psi(x) = \sin\,(k_0 n x + \phi + \vartheta) \qquad \text{S8}$$

Where ϕ is given by Eq. S5.i and $\vartheta = e^{-2}k_0\kappa_m t_m$ being equal to the photonic tunneling probability as calculated elsewhere.[1] The solutions for such a "leaky" double-potential-well are, therefore:

$$\tanh\left(k_0\kappa_m \frac{t_{cm}}{2}\right) = \frac{\kappa_m}{n_d} \tan\,(-k_0 n_d \left(\frac{1}{k_0\kappa_m} + t_d\right) - \exp\,(-2k_0\kappa_m t_m)) \qquad \text{S9}$$

$$-\coth\left(k_0\kappa_m \frac{t_{cm}}{2}\right) = \frac{\kappa_m}{n_d} \tan\,(-k_0 n_d \left(\frac{1}{k_0\kappa_m} + t_d\right) - \exp\,(-2k_0\kappa_m t_m)) \qquad \text{S10}$$

These solutions are valid in the case of normal incidence. In the main manuscript, however, a more general solution in which the angular dependence is included was needed to model the behavior shown in Figures 2c and 4e. Straightforwardly, for a p-polarized wave impinging with a sufficiently high angle like 40°, Equation S9 and S10 can be readily modified considering that the tunneling coefficient through the metal has to be divided by the sinθ:

$$\tanh\left(k_0\kappa_m \frac{t_{cm}}{2}\right) = \frac{\kappa_m}{n_d} \tan\,(-k_0 n_d \left(\frac{1}{k_0\kappa_m} + t_d\right) + \left(\frac{1}{\sin\,(\theta)}\right) \exp(-2k_0\kappa_m t_m)) \qquad \text{S9}$$

$$-\coth\left(k_0\kappa_m\frac{t_{cm}}{2}\right)=\frac{\kappa_m}{n_d}\tan\left(-k_0n_d\left(\frac{1}{k_0\kappa_m}+t_d\right)+\left(\frac{1}{\sin(\theta)}\right)\exp(-2k_0\kappa_m t_m)\right) \quad \text{S10}$$

Bibliography

[1] Caligiuri V, Palei M, Biffi G, Artyukhin S, Krahne R, A Semi-Classical View on Epsilon-Near-Zero Resonant Tunneling Modes in Metal/Insulator/Metal Nanocavities. Nano Lett 2019, DOI: 10.1021/acs.nanolett.9b00564.