Supplementary Information

Omnidirectional absorption in nanostructured metal surfaces

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General conditions for total light absorption

We find it appropriate to use a Hamiltonian formalism to recast Maxwell's equations in the form of Schrödinger equation. Then, the general principles of formal scattering theory developed in the context of quantum mechanics can be applied to analyze the interaction of light with resonant nanostructures [1,2].

The solution of our scattering problem (i.e., light reflection on a nanostructured surface tailored in an opaque substrate) can be written in the form of Lippmann-Schwinger equation [3]:

$$\Psi = \Psi_0 + G^+(\omega)(H - H_0)\Psi \equiv \Psi_0 + \Psi^+, \tag{1}$$

where ω is the light frequency, acting here as the *quantum* energy, H and H_0 are Hamiltonians representing structured and planar metal surfaces, respectively, Ψ^+ is an outgoing wave function containing all components of electric and magnetic fields for the structured surface, Ψ_0 is the wave function of the *unperturbed* planar-surface system that satisfies $(H_0 - \omega)\Psi_0 = 0$ and corresponds to specific angle of incidence and polarization, and $G^+(\omega)$ is the retarded Green function of H.

The whole energy flux in the system is distributed in different *quantum* channels, among which we single out the *elastic* one (channel 1) describing the incident light and its specular reflection without polarization conversion. Other channels include specular reflection with polarization conversion and diffracted beams.

Neglecting absorption in the flat metal surface, the asymptotic magnetic field of channel 1 can be written

$$\mathbf{B}_1 = \frac{1}{\sqrt{k_\perp}} \left[\hat{\mathbf{e}}_i \exp(-ik_\perp z) + \hat{\mathbf{e}}_r S_{11} \exp(ik_\perp z)\right], \tag{2}$$

where $\hat{\mathbf{e}}_{i,r}$ defines the polarization direction for incident (i) and reflected (r) waves, and $k_\perp$ is the wave vector component perpendicular to the structure (along the direction z). The first term in Eq. (2) represents the incident light, whereas S_{11} is the scattering amplitude. In the vicinity of a pole associated with a localized resonance, the Breit-Wigner multichannel scattering theory permits

approximating S_{11} by [4]

$$S_{11} = \exp(2i\delta_{11}) - \frac{iM_{11}\Gamma}{\omega - \omega_0 + i\Gamma/2}\exp(2i\delta_{11}), \tag{3}$$

where the second term describes resonant scattering. Here, ω_0 and Γ are the frequency and width of the resonance, respectively, δ_{11} is a non-resonant scattering phase, and M_{11} is a constant.

Flux conservation in the system implies

$$|S_{11}|^2 + J = 1, \tag{4}$$

where J is the flux associated to channels other than 1. In what follows we assume that the partial decay rate into channel 1 (i.e., the relevant *radiative* decay rate) equals the sum of rates into all other channels:

$$J = \left|\frac{M_{11}\Gamma}{\omega - \omega_0 + i\Gamma/2}\right|^2. \tag{5}$$

Now, using Eqs. (3), (4), and (5), we obtain

$$|M_{11}|^2\Gamma = Re\{iM_{11}(\omega - \omega_0) + M_{11}\Gamma/2\}. \tag{6}$$

Equation (6) should hold for all frequencies, so that $M_{11} = 1/2$, and the specular reflection coefficient (reflection into channel 1) reduces to

$$R = |S_{11}|^2 = \frac{(\omega - \omega_0)^2}{(\omega - \omega_0)^2 + \Gamma^2/4}. \tag{7}$$

Clearly, R drops to zero at the resonance frequency $\omega = \omega_0$, at which the incident flux is completely transferred into polarization conversion, diffraction, and absorption.

This allows us to assess the conditions for total resonant light absorption assisted by a localized resonance as follows: (i) there is only specular reflection with no diffracted beams; (ii) there is no polarization conversion; and (iii) the radiative decay rate of the resonance equals its dissipative decay rate (rate equipartition condition). Under these conditions the incident light is fully transformed into losses in the metal, such that the absorption $A = 1 - R$ is 100% at the resonance frequency $\omega = \omega_0$.

Interaction between surface-plasmon polaritons and localized plasmons

The total absorption effect that we are discussing, which is assisted by localized plasmons, can be damaged by interaction with delocalized surface plasmons of the planar surface above the voids, as shown in Fig. S1. This interaction has been considered in recent literature [5,6], and we show it here under the specific conditions of our study (see Fig. 1 in the main text).

The absorption is dominated by interaction with two types of plasmons: modes localized at the voids, which produce angle-independent absorption features, and delocalized planar-surface plasmons. These two types of modes can be relatively close in frequency, as shown in Fig. S1, where an avoided crossing is clearly observed, involving hybridized plasmons of mixed character [5,6]. The interaction becomes apparent for angles of incidence above 40°, where absorption omnidirectionality is destroyed.

This problem is solved in the main text by filling the voids with a dielectric, so that the void plasmon is brought down in energy to a region where it does not interact with planar-surface

plasmons (see Fig. 2).

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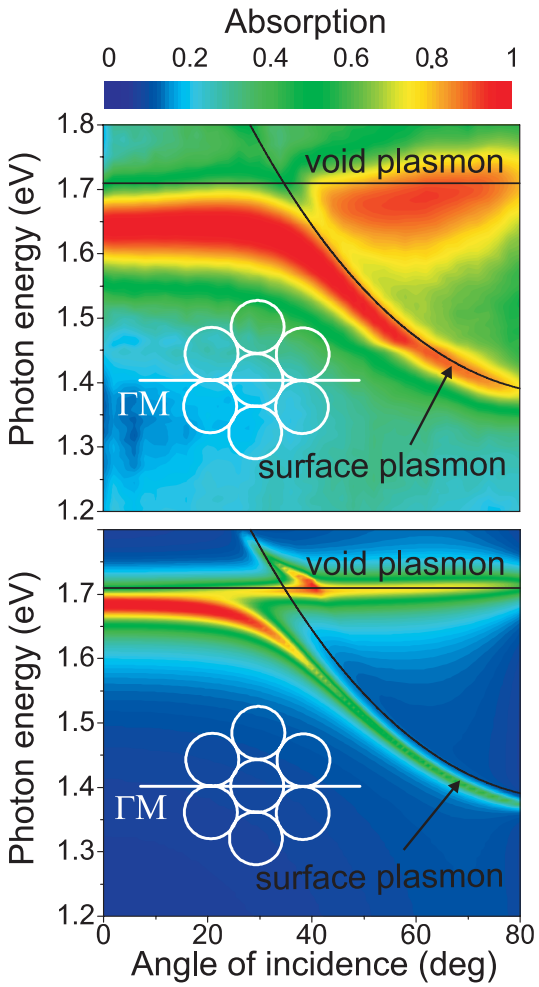

Figure S1. Measured (top) and calculated (bottom) absorption of samples considered in Fig. 1A, represented for p-polarized light incident along the $\Gamma - M$ direction of the void lattice (see insets). The thickness of the nanoporous layer ($t = 1.08$ and $t = 1.024$ times the void diameter, respectively) is chosen to realize the total light absorption condition (i.e., the rate equipartition) at normal incidence. The surface plasmon folding over the void lattice and the dipolar single-void plasmon mode are shown as black curves.