Supporting Information


Ultrathin Semiconductor Superabsorbers from the Visible to the Near-Infrared

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1. **Resonant modes sustained by a flat thin film**

![Figure S1. Scheme of the studied multilayer system used to obtain the dispersion relations of the layered structure.](image)

We consider herein the simple case of a three-layer media illustrated in the **Figure S1**. This system can be understood as an asymmetric waveguide coupled to a resonator or cavity. To determine the condition of total absorption for this layered system, we must find the condition for critical coupling of the waveguide to the resonator\[^1\], i.e., no reflection to the input and no transmission to the output of the waveguide. This becomes possible by perfect coupling of incident light into the optical modes of the resonator (or equivalently, by a destructive interference of light in the outside medium). Incident light can be completely trapped in the resonator, and eventually be totally absorbed. It was found\[^2\] that this critical coupling occurs when the internal dissipation rate in the resonator is equal to the leakage (or radiative losses) rate of the waveguide.

In this way, we consider an a-Ge planar waveguide of thickness $t$ with permittivity $\varepsilon_2$ surrounded by two semi-infinite media (air and gold) with dielectric permittivities $\varepsilon_1$ and $\varepsilon_3$, respectively (**Figure S1**).
For TM polarization (magnetic field transversal to the incidence plane) the scattered electric fields are

\[ E_1 = \left( ik_{\|}, 0, -\frac{k_{\|}^2}{a_1} \right) E_1 \exp(-a_1 z) \]  
(1)

\[ E_2 = \left( ik_{\|}, 0, \pm\frac{k_{\|}^2}{a_2} \right) E_2 \exp(\pm a_2 z) \]  
(2)

\[ E_3 = \left( ik_{\|}, 0, \frac{k_{\|}^2}{a_3} \right) E_3 \exp(a_3 z) \]  
(3)

Where the attenuation constants are

\[ a_i^2 = k_i^2 - k_0^2 \varepsilon_i \quad \text{for} \quad i = 1, 2, 3. \]  
(4)

Applying the appropriate boundary conditions at the two interfaces gives the dispersion relation for the surface waves propagating parallel to the surface\(^3\)

\[ \frac{a_2 \varepsilon_3}{a_3 \varepsilon_2} + \frac{(a_2 \varepsilon_1 + a_1 \varepsilon_2) + (a_2 \varepsilon_1 - a_1 \varepsilon_2) \exp(-2a_2 t)}{(a_2 \varepsilon_1 + a_1 \varepsilon_2) - (a_2 \varepsilon_1 - a_1 \varepsilon_2) \exp(-2a_2 t)} = 0. \]  
(5)

When \( t \gg \lambda \) in Equation 5, we recover the well-known surface plasmon polariton equation for a dielectric-metal interface:

\[ k_{\|}^2 = k_0^2 \varepsilon_2 \varepsilon_3 / (\varepsilon_2 + \varepsilon_3) \]  
(6)
In the case of lossless materials it is easy to see that, if \(-Re(\varepsilon_2) < Re(\varepsilon_3) < 0\), then \(k_\parallel\) is purely imaginary. This means that the modes correspond to non-propagating waves or localized modes\(^4\).

The energy of the evanescent surface wave is dissipated into two radiative waves propagating away from the surface. When we introduce losses in the system, \(k_\parallel\) becomes complex with a high imaginary part and the two waves become evanescent. As explained in the main text these conditions relates to a quasi-bound regime.

The previous permittivity relation occurs for amorphous germanium on a gold substrate, up to a wavelength \(\lambda = 764\, \text{nm}\), and the introduction of losses for gold and germanium favours high values for the imaginary part of \(k_\parallel\) in a broader bandwidth.

For the TE polarization in nonmagnetic media (electric field transversal to the incidence plane) taking the magnetic permeabilities as \(\mu_1 = \mu_2 = \mu_3 = 1\); the dispersion relation is given by

\[
\frac{a_2}{a_3} + \frac{(a_2 + a_1) + (a_2 - a_1)\exp(-2a_2t)}{(a_2 + a_1) - (a_2 - a_1)\exp(-2a_2t)} = 0,
\]

although the surface wave is not permitted at an interface dielectric/metal for this polarization\(^5\), when an ultrathin film of high dielectric permittivity is inserted on the metal substrate the existence of an artificial surface wave in nonmagnetic media has been reported.\(^6\)

Solving Equation 5 and 7 imply that we must consider either \(k_\parallel\) or \(\omega\) to be complex. We prefer here the option where \(\omega\) is real and \(k_\parallel\) complex, since it is better adapted to situations with continuous wave excitations. A complex \(k_\parallel\) is a way to represent a mode that decays as it propagates away from the region where the external excitation was applied. In practical situations, it is possible to recover the case where \(k_\parallel\) is real by assuming that \(\omega\) is complex, to account for the fact that the modes have a finite lifetime.\(^4\)
In Figure S2 we can observe the evolution of the absorption profile of a 15 nm thick flat a-Ge film on gold as we change the angle of incidence. In agreement with previous reports\textsuperscript{[7,8]}, we find that such a simple layered configuration can exhibit coupling between the incident light and a bound Brewster-type resonance whereby the energy of an incident plane wave is almost totally absorbed by the active medium. These Brewster modes are radiative modes which become surface modes in the presence of losses.\textsuperscript{[4]} The characteristic width of these absorption peaks originates in the above-mentioned quasi-bound regime of high damped modes where the reflected field is entirely suppressed.

The mechanism for this effect resides in the strong interaction between the semiconductor layer and the metal substrate that occurs when the incident wave is phase-matched to the radiated field supported by either layered media. The large confinement factor together with a slow group velocity due to the high refractive index make the system a candidate for high absorption.\textsuperscript{[9]} Our simulations indicate that total absorption of the incident energy may occur in both TE and TM polarization, as Figure S2a and b show, respectively. We calculated with FDTD the dispersion curves of the system shown in Figure S2d. It is shown that for a thin layer of 15 nm the dispersion relation for the TM polarization reaches values of $k_{\parallel} \approx 0$ i.e., can be excited from normal incidence.
**Figure S2.** Total absorption versus wavelength and angle of incidence in a 15 nm thick Ge layer for TE a) and TM polarizations b). c) and d) dispersion relations (frequency versus the parallel wavevector) for TE and TM polarizations, above the light line (k_∥ < k_0) showing the radiation modes which are the accessible modes from the external incident light. For k_∥ > k_0 the dispersion (green dashed curve) of the system calculated by FDTD is shown.

When the thickness of the active layer is increased, the absorption band due to the strong interference shifts to longer wavelengths. At these wavelengths, gold behaves closer to an ideal metal and $Re(\varepsilon_3) < -Re(\varepsilon_2) < 0$. This characteristic, together with the low values of the absorption coefficient of Ge for these wavelengths, results in a much lower absorption for the NIR range. This situation is illustrated by comparing **Figure S3** for a layer of 70 nm thickness and **Figure S2** for a 15nm thick layer. Also, the dispersion relation of the system for both polarizations is shown for the non-accessible wavevectors $k_∥ > k_0$. In this case a guided mode appears for the TE polarization. For the TM polarization the dispersion relation in the quasi-bound mode reaches
only $k_{||}$ close to the light line due to the increasing thickness\cite{10}, as it is corroborated by the absorption profile in this zone. Therefore light at normal incidence cannot be coupled to this mode, substantially decreasing the absorption for normal incidence.

![Figure S3. Total absorption versus wavelength and angle of incidence in a 70 nm thick Ge layer for TE a) and TM polarizations b). c) and d) dispersion relations (frequency versus the parallel wavevector) for TE and TM polarizations, above the light line ($k_{||} < k_o$) showing the radiation modes which are the accessible modes from the external incident light. For $k_{||} > k_o$ the dispersion (green dashed curve) of the system calculated by FDTD is shown.](image)
2. *Pillar and hole array structure discussion*

In Figure S4 we show the absorption profiles of the two possible square lattice arrays: a hole array structure versus a pillar array structure with the same structure parameters. Those comprise a thicker thickness $t_2$ and a thinner thickness $t_1$ of 70 nm and 15 nm respectively, a lattice parameter L=700 and the same radius for both the holes and pillars of 300 nm. Two different absorption profiles are obtained reflecting the different interacting modes supported by each structure but reaching similar integrated absorptions of 60.8% and 60.5% from 400 nm to 1500 nm for holes and pillars respectively. However the hole array structure presents a 14% more absorption in the 1000 - 1500 nm range while the pillar array has a slightly better absorption in shorter wavelengths.

![Figure S4. Total (dashed lines) and semiconductor (solid lines) absorptions of a hole array photonic structure (orange) and a pillar array structure (black) with $r = 300$ nm, $t_1 = 15$ nm, $t_2 = 70$ nm and L = 700 nm.](image-url)
3. **Angle of incidence and polarization behaviour of the structure surface.**

In **Figure S5** we show the absorption behaviour of the 2D Ge PhC analysed in the main text as a function of wavelength, polarization and angle of incidence. The Brewster resonant mode excited at visible wavelengths spatially located in the thinner ($t_1$) 15 nm layer shows high absorption for all angles in TM polarization while maintaining it up to 70% until 45° with TE polarization. For longer wavelengths, in the NIR region, we observe high absorption flat bands for both polarizations even for large angles up to the first diffraction order line (See black dotted line in **Figure S5c and d**). Above the diffraction line, at higher angles, there are radiation losses because part of the light is diffracted back to air.

As we have noted in the main text, guided modes cannot be excited from outside an unpatterned thin film because of insufficient $k_\parallel$. Using periodic structures can be beneficial to enhance light trapping, for instance a bidimensional grating can provide access to $k_\parallel > k_\sigma$. The parallel wavevectors $k_\parallel$ can be obtained from Equation 1 of the main text where $[b_x, b_y] \neq [0,0]$ gives the diffraction order inside the semiconductor layer. In addition, the decomposition of radiation phenomena into a superposition of discrete modes is a powerful concept; if combined with periodic boundary conditions, each discrete mode can be identified by a point in an equidistant reciprocal lattice. This is schematically drawn in **Figure S5c** for normal incidence where the plasmon TM and the photonic guided TE modes of a flat layer, obtained with an effective medium theory at second order[11], are represented (green curves) together with the coupling with the different grating orders. This shows a good agreement with the position of the NIR peaks and even the order $[\pm2,0]$ and $[0,\pm2]$ coupled with the TE guided mode fits with the small absorption peak in $\lambda =1000 \text{ nm}$. 

Figure S5. Total absorption versus wavelength and angle of incidence in a photonic structure with $r = 300\, \text{nm}$, $t_1 = 15\, \text{nm}$, $t_2 = 70\, \text{nm}$ and $L = 700\, \text{nm}$ for TE a) and TM polarizations b). Dispersion relation of frequency versus parallel wavevector for both polarizations, c) and d), respectively. Scheme of the decomposition of radiation phenomena into a superposition of discrete modes, c). The vertical blue lines correspond to the $k_{\parallel}$ of the diffraction orders of the lattice which couple to the modes above the light line (green dotted lines).
4. Electric and magnetic field distributions of an amorphous germanium film with a square array of holes.

In Figure S6 the simulated magnetic and electric field intensities are larger in the thicker layer $t_2$ in correspondence with the peaks shown in Figure 4 of the main text. Also, the hybridization of two surface waves, one located at the interface air/Ge and a second at the Ge/Au interface, can be observed.

![Image of electric and magnetic field distributions](image)

$\lambda = 1170$ nm

$\lambda = 1370$ nm

**Figure S6.** Intensity of the electric and magnetic field at the plane shown in the top inset at wavelengths, 1170 and 1370 nm for the nanostructure considered in the main text (the units are normalized to the maximum of each plot).

This indicates that the inclusion of the metal substrate plays an important role both for the interference resonance and the large confinement. We can enlighten this phenomena representing the field behaviour of the a-Ge photonic structure without metal substrate and comparing with the complete system metal/semiconductor (Figure S7). The confinement is significantly larger in the structure with metal substrate.
Figure S7. Effect of the metallic substrate in the confinement of the electric field. a) absorption at normal incidence of the system calculated by FDTD for the a-Ge hole square array over metal (blue solid line) and without metal (red dashed line). b) Scheme of the cross section of the structures perpendicular to the plane of incidence for TM polarization. Intensity of the electric field at the resonant wavelengths for the nanostructure considered in the main text without c), e) and with the gold substrate d) and f). The resonant wavelengths are at 1035 nm c), 1240 nm e), 1170 nm d) and 1370 nm f).
5. **Tuning the absorption profile of the photonic structure with the geometrical parameters.**

As we have seen in the main text, the resonances at long wavelengths show a strong dependence with the geometric parameters of the photonic architecture. Here we elaborate on the versatility of our structure.

In **Figure S8** we represent the absorption of an architecture where the hole radius \( r \) is varied in the array structure for a fixed lattice parameter \( L = 700 \, \text{nm} \). The structure evolves from a flat 70 nm thick Ge layer (I) \((r = 0)\) to a flat 15 nm thick Ge layer (VI) \((r = \sqrt{2}/2 \, L)\), by opening holes. For radius reaching \( r \approx 100 \, \text{nm} \) (II) the photonic modes due to the structured layer begin to appear. The separation between these resonances increases with the radius (III). Furthermore, the increase of the radius involves a decrease of the fill factor of \( t_2 \) vs. \( t_1 \) and the absorption due to the strong interference in the thinner layer of \( t_1 = 15 \, \text{nm} \) emerges in the visible range. For \( r = \frac{L}{2} = 350 \, \text{nm} \) the holes start to overlap each other tangentially (IV) and the peak with the electric field confined at this position (absorption peak II of **Figure 4** of the main text) disappears. The same happens with the mode between four holes, in their cell diagonal, as the inverse cell structure decreases (V) this absorption decreases and shifts to shorter wavelengths (absorption peak III of **Figure 4** of the main text).
Figure S8. Evolution with radii of the calculated total absorption versus wavelength at normal incidence of the photonic architecture with $L = 700$ nm, $t_1 = 15$ nm and $t_2 = 70$ nm.

The thicknesses $t_1$ and $t_2$ are also essential characteristics in order to control the spectral positions of the absorption peaks as illustrated in Figure S9 for both the total absorption and the fraction absorbed in the Ge active layer. The increase of $t_1$ when $t_2 = 70$ nm is fixed (Figure S9a and b) causes an increase of the position in wavelength of the Brewster mode confined in the thinner slab $t_1$ whereas the other peaks associated to $t_2$ remain fixed. Furthermore, in Figure S9c and d thickness $t_1 = 15$ nm is fixed and thickness $t_2$ is varied. We can observe that for increasing $t_2$ the associated absorption peaks with the electric fields localized in $t_2$ shift towards longer wavelengths whereas the peak associated to the thinner thickness $t_1$ remains fixed.
Figure S9. **Evolution of the total absorption a) and a-Ge absorption b) in a square hole array with 700 nm lattice parameter and 300 nm radius. The larger thickness $t_2$ is kept fixed at 70 nm and the smaller thickness $t_1$ inside the holes is varied. Total absorption c and a-Ge absorption d) in a square hole array with 700 nm lattice parameter and 300 nm radius. The larger thickness $t_2$ is varied and the smaller thickness $t_1$ is kept fixed at 15 nm.**

In Figure S10, we show extinction measurements for several samples that correspond to photonic crystal structures. In a, c, e the lattice parameter and the hole diameter are 700 nm and 560 nm respectively, and the larger a-Ge thicknesses $t_2$ are 60, 70 and 80 nm, as indicated. The observed changes are in agreement with the calculations described above. The same trend is found in the graphs b, d and f, for spectra measured on photonic crystals with 1100 nm lattice parameter and 880 nm hole diameter. In addition, for every fixed a-Ge thicknesses, larger photonic crystal lattice parameters are shown to effectively shift resonant modes towards longer wavelengths.
**Figure S10.** Experimental extinction of the designed structure varying both layer thickness and lattice parameter showing tuneable absorption. Three thicknesses have been used: 60, 70 and 80 nm, one for each row of plots. Two parameter configurations are shown: the first column set samples had a lattice parameter $L = 700$ nm with a diameter $D = 560$ nm and the second column samples had the parameters $L' = 1100$ nm and $D' = 880$ nm.

6. **Refractive index of amorphous germanium obtained by ellipsometry.**

The a-Ge optical constants and the planar film thicknesses were obtained from analysis of ellipsometry measurements. In the present study, the optical constants were determined from a film nominally 60 nm thick. The measured ellipsometric quantities at several angles of incidence $\varphi$ compared to the fittings from which $n$ and $k$ were deduced are plotted in **Figure S11a and b**. The parametric representation allows extrapolating the $n$ and $k$ values to longer wavelengths beyond the spectral range of the used ellipsometer. These values are plotted in **Figure S11c** where the obtained
parameters for the Cody-Lorentz\cite{12} dispersion are: $\varepsilon(\infty) = 0.60$, $A = 86$ eV, $E_0 = 3.49$ eV, $\Gamma = 4.33$ eV, $E_g = 0.80$ eV, $E_p = 0.74$ eV, $E_t = 1.22$ eV, and $E_u = 0.193$ eV, corroborated with FTIR measurements.

**Figure S11.** Measured ellipsometric quantities (symbols) at several angles of incidence $\phi$ compared to the calculated values (lines),  
(a) $\tan \psi$, (b) $\cos \Delta$. The number of measured points has been reduced for clearer representation purposes. The sample was modelled as a film with a Cody-Lorentz dispersion on Au substrate. A surface roughness layer was included in the analysis. The thicknesses obtained from the fits were $59.8 \pm 0.5$ nm (film) and $4.8 \pm 0.5$ nm (roughness), equivalent to a total deposited Ge thickness of 62.2 nm.  
(c) optical constants of the MBE-deposited a-Ge film fitted from ellipsometry.
Supporting information References


