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ABSTRACT

Scanning-probe-assisted mid-infrared nano-spectroscopy is employed to reveal the polaritonic dispersion of individual MIM (metal-insulator-metal) square patch antennas whose modes can be strongly coupled to a mid-infrared intersubband transition. The patch antenna side length L sets the resonances between $k = 5.5\,\text{nm}$ and $12.5\,\text{nm}$. The active region consists of a highly doped AlInAs/InGaAs/AlInAs single quantum well that presents an intersubband transition at $1190\,\text{cm}^{-1}$ ($\omega = 8.4\,\text{meV}$). When the patch antenna optical resonance approaches and matches the intersubband transition frequency ($L = 1.8\,\text{nm}$), a clear anticrossing behavior—evidence of strong coupling—is observed in the near-field scattering phase spectra of individual antennas. The measured Rabi splitting is $4.5\,\text{THz}$. The near-field scattering spectra agree with the far-field extinction spectra acquired on arrays of identical antennas.

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Intersubband (ISB) transitions in semiconductor quantum wells (QWs) enable the implementation of devices operating in the mid-IR (and THz) spectral ranges, such as the quantum cascade laser1 and the quantum well infrared photodetector.2 Furthermore, they are important in the study of fundamental physical phenomena at very long wavelengths, such as non-linearities, cavity electrodynamics (CED), and strong light-matter coupling. In the latter regime, the light-matter coupling is stronger than the dephasing mechanisms in the system, new quasi-particles—called ISB polaritons—emerge.3,4 ISB polaritons have been subject to much theoretical and experimental work since their first observation, with developments including ISB polariton LEDs,5,6 polariton-based detectors,7 ultra-fast switching,8 and ultra-strongly coupled systems.9,10

Recently, the study of ISB polaritons at the single resonator level has started to attract interest. This partly stems from experience in the visible and near-infrared spectral ranges, where it is possible to operate on isolated resonators with very high Q/V values (Q is the quality factor and V is the resonator modal volume).11 This has been, however, challenging in the mid-IR and THz spectral ranges because the detectors are less efficient, among other factors. With standard far-field techniques, only ensembles of a few cavities can be measured.12,13 Remarkably, using thermal fluctuations as an internal source of electromagnetic field, a single patch cavity could be studied,14 but a temperature of 433 K was necessary, which is possibly not compatible with strong light-matter coupling.

Operating in the electromagnetic near-field offers an alternative way to access the optical properties of a single resonator at very long wavelengths. Near-field scattering optical microscopy (SNOM) techniques, based on scanning probes approaching the object, have recently been exploited to study empty cavities15 and systems operating in the weak coupling regime.16-19 The challenge when moving to the strong-coupling regime is to maintain an elevated electromagnetic overlap...
with the active region—to obtain a large Rabi splitting—while permitting a non-negligible fraction of the electric field to leak outside the cavity in the form of “fringing” fields that can be scattered by the scanning probe.

In mid-IR/THz polaritonics, metallic cavities, typically in a metal-insulator-metal (MIM) architecture, are almost exclusively used, with the patch antenna proving the most effective in terms of electromagnetic confinement, tailorable radiative coupling,22,23 and easy implementation in more complex devices. Its extreme confinement properties explain why there is no report of near-field field spectroscopy on patch antennas strongly coupled to ISB transitions in QWs, although an initial demonstration has been very recently given in dogbone antennas coupled to ISB transitions.24

In this Letter, we show that judicious design and careful experimentation permit broadband SNOM nanospectroscopy to be performed on a single patch antenna strongly coupled to a mid-IR ISB transition: the polaritonic resonances appear in the near-field scattering phase spectra of single antenna cavities, indicating that strong coupling is achieved in individual resonators and antenna arrays are not required.

In our MIM antenna, the two mirrors of the optical cavity are formed by a top gold patch (square of side L) and a bottom gold backplane. The cavity contains a dielectric layer (polymer, inserted for the purpose of photothermal expansion imaging, see below) and a heterostructure semiconductor (a heavily n-doped multisubband QW).

The electromagnetic simulations, performed with an FDTD (finite-differences time-domain) approach [Fig. 1(a)], reveal a strong field enhancement (Ez/Einc) in a patch antenna with its cavity filled with a 200 nm-thick dielectric layer and a 120 nm-thick semiconductor layer containing a QW. The cavity contains a dielectric layer (polymer, inserted for the purpose of photothermal expansion imaging, see below) and a heterostructure semiconductor (a heavily n-doped multisubband QW).

The final realization of a typical sample is represented in Figs. 1(b) and 1(c). The two-level system is constituted by a single highly doped QW [AllnAs 25 nm/InGaAs 18.5 nm/AllnAs 25 nm, Fig. 1(d)], exhibiting a multisubband ISB transition in the mid-IR at 1190 cm−1 [E2 − E1 = 148 meV, Fig. 1(e)]. In the fabrication process, the active region was coated with a metallic layer (10 nm Ti/100 nm Au, e-beam evaporated) and subsequently bonded to a clean GaAs host wafer with a commercial epoxy adhesive (Epo-tek 353-ND, Epoxy Technology). The pristine InP growth substrate was then removed in HCl, exposing the InGaAs etch-stop layer (40 nm) above the QW structure. A 200 nm-thick polymeric layer (polymerized EPON 812 by Sigma Aldrich) was cut with an ultramicrotome (Leica EM UC7) and transferred—exploiting the high surface tension of distilled water, which keeps the thin epilayer unfolded and stretched—onto the semiconductor active region.25 A 100 nm-thick gold layer was finally evaporated on top of the resist. The final step was the implementation of patch antennas in layouts of either arrays (between 100 and 150 elements, depending on the value of L; periodicity in arrays p = 2L) or of single isolated cavities, using focused ion beam patterning to etch away the unwanted gold. A few tens of nanometers of polymer were etched and removed as well, without, however, affecting the optical response.

The arrays were used to measure far-field extinction spectra using unpolarized light with a Fourier transform IR spectroscopy (FT-IR) microreflectance setup in the 600–4000 cm−1 range (Nicolet Continuum), equipped with a 32×, 0.65 NA Cassegrain objective.

As stated previously, access to a scattering-type SNOM of the patch antenna cavity, where the field is confined, is a problem. Near-field imaging of the mode profile cannot be performed by scanning the patch antenna surface, as the field is practically zero above the top metallization [Fig. 1(a)]. To access the field distribution in an alternative way, we, thus, explored photothermal expansion microscopy to obtain a sub-diffraction-limit-resolved image of the patch antenna cavity. In this technique, a monochromatic IR laser beam is focused on a scanning probe (similar to SNOM), but instead of the scattered radiation, the mechanical atomic force microscopy (AFM) signal is acquired. The local thermal expansion is measured by optomechanical analysis of the AFM cantilever oscillations at the repetition rate of the IR laser (AFM-IR technique).25-27 In our case, radiation energy absorption is sensed at a specific xy position inside the antenna cavity by determining the IR-induced thermal expansion at that position, with the probe tip scanning the surface of the gold patch. In order to enhance this AFM-IR signal and the imaging resolution, a 200 nm-thick layer of EPON 812 polymer (that features a high thermal expansion coefficient and a poor lateral thermal conductivity) is inserted into the antenna cavity during sample fabrication. The AFM-IR measurements were performed using a NanoIR2 (Anasys Instruments), using the same configuration employed in SNOM measurements: a gold-coated probe tip, 70° incidence of the IR laser beam, and the radiation electric field linearly polarized in the incidence plane (p-polarization).

The spectra of the IR radiation scattered by single antennas were collected with a SNOM microscope (NeaSNOM from NeaSpec). Linearly polarized broadband IR radiation, obtained from difference-frequency generation, is focused on the gold-coated AFM probe tip, operating in the tapping mode, through a parabolic mirror that is also used to collect the backscattered radiation. The detected signal is amplitude-modulated at the mechanical tapping frequency ~220 kHz. Because of the non-linear dependence of the near-field interaction from the tip-to-sample distance, at higher harmonics n (with n ≥ 2),
the near-field signal from the tip-antenna interaction is dominant over the
linear far-field background. The scattering spectra are extracted
using the nano-FTIR technique in which the tip and the sample are in
an arm of a Michelson interferometer, thereby providing both the
amplitude and phase of the backscattered radiation. The spectra
were taken with the probe tip located at the bottom right antenna cor-
nor on which the light impinges first, where the highest signal intensity
was observed [see Fig. 2(c), X mark]. The measurements were taken
by averaging 10 interferograms with a spectral resolution of 8 cm−1. The
demodulated phase and amplitude signals are normalized to the
reference quantities measured on an Au film deposited far from the
antennas,

\[ \eta_n(\omega) = s_n^{\text{norm}} e^{i\phi^{\text{norm}}} = \frac{s_n^{\text{antenna}}}{s_n^{\text{g}}} e^{i(\phi^{\text{antenna}} - \phi^{\text{g}})}, \tag{1} \]

where \( s \) is the near-field amplitude, \( \phi \) is the phase, and the suffix \( n \)
indicates the \( n \)th harmonic demodulation used to suppress the far-
field background. In the case of absorbing materials, in the limit of
sample thickness larger than the inverse characteristic value of the in-
plane momentum excited by the probe tip, it has been shown that
the far-field absorption peak positions correspond to those in the
imaginary part of the SNOM signal: \( \text{Im}(\eta_n) = s_n^{\text{norm}} \sin(\phi_n^{\text{norm}}). \) For
thinner samples, the peak position in the scattering phase (\( \phi^{\text{norm}} \)) or
in \( \text{Im}(\eta) \) corresponds to features typically observed in grazing-
incidence IR reflection spectroscopy with p-polarized light. For complex
structures such as antennas, the scattering coefficient will be the
result of the electromagnetic interaction of the tip with the antenna
(in our case, also coupled to the QW transition) and with the substrate.
This interaction can be described either as coupled harmonic
oscillators or as a set of complex optical impedances. As a result,
the SNOM observations of complex tip-antenna systems strongly vary
in the literature depending on the relative interaction strength of
the antenna and the tip with the exciting radiation and the relative delay
of this interaction, resulting in a dip in the SNOM signal amplitude, a
dispersive step-like behavior in the phase, a peak in the imaginary
part, or a step-like feature in the imaginary part.17

Experimentally, we observed peaks in \( \phi^{\text{norm}} \) [Fig. 2(a)] as well as in
\( \text{Im}(\eta) \) (not shown). The peaks in \( \text{Im}(\eta) \) are always red-shifted,
within the width-at-half-maximum of the \( \phi^{\text{norm}} \) peak for all \( L \), and so
analyzing \( \text{Im}(\eta) \) instead of \( \phi^{\text{norm}} \) would not impact the following
discussion.

The near-field scattering spectra in Fig. 2(a) exhibit a clear anti-
crossing behavior, with a minimum splitting obtained for \( L = 1.9 \mu m \).
In this case, the full width at half maximum for both polaritons is
found to be 195 cm−1. This value is approximately equal to the splitting
energy (205 cm−1) and indicates a strong-coupling regime, further
corroborated by comparison with the extinction measurements
performed in the far-field with a FTIR microscope on patch antenna
arrays [Fig. 2(b)]. The range of \( \eta \)-values where the strong coupling is
observed, and the polariton peak positions and their relative peak
intensity, as measured with the two techniques (near-field vs far-field),
are similar. However, for \( L < 1.5 \mu m \), where the detuning is large and
only one uncoupled-antenna peak is observed, the near-field (SNOM)
peak is slightly blue-shifted when compared to the far-field (FTIR)
peak.31,32 The polariton dispersion is highlighted by the same lines
that are guides to the eye in both Figs. 2(a) and 2(b) (dashed black
curves). The data in Fig. 2(a) are a clear indication of strong coupling
between isolated individual antenna resonators, such as
the patch antenna in Fig. 1(c). A peak located at 1738 cm−1, corre-
spending to the C=O stretching vibration in the EPON polymer, is
visible in the FTIR spectra [Fig. 2(b)] and couples also with the plas-
mon resonance of the patch antenna for \( L = 1.3–1.2 \mu m \), leading to an
asymmetric resonance and a surface-enhanced infrared absorption
(SEIRA) effect.

In Fig. 2(a), with \( L = 2.2 \mu m \) and \( L = 2.3 \mu m \), the shoulder of a
high frequency peak can be seen (not fully shown in the graph for clar-
ity). This peak is attributed to the second-order antenna resonance,
which couples very strongly to the probe. The AFM-IR maps in
Figs. 2(c)–2(e), taken far from strong coupling for clarity (\( L = 2.2 \mu m \)
and \( L = 1.2 \mu m \)), show that a single antenna can be correctly excited
by a gold-coated AFM probe when the wavelength is resonant with
the antenna mode, the incidence angle is 70°, and the radiation
electric-field polarization is TM (p-polarized wave), like in the
SNOM experiment.

In Fig. 3, we summarize the SNOM nanoscopy results (red stars) and compare them to far-field spectroscopy (blue dots). Both series of data points have been obtained by fitting of the spec-
tra in Fig. 2 with Lorentzian functions that reproduce the main
spectral features. The analytical prediction for strong coupling
based on the secular equation is shown as a continuous green line,12
which are the roots of the following secular equation that is
an implicit equation for \( \omega_f \):

\[
\left( \omega_f^2 - \omega_{\text{patch}}^2 \right) \left( \omega_f^2 - \tilde{\omega}_{\text{ISB}}^2 \right) = \Gamma_{\text{opt}} f_w \omega_{\text{patch}} \omega_{\text{plasma}}^2.
\]

These roots correspond to the frequencies of upper and lower
and polaritons. \( \omega_{\text{patch}} \) is the frequency of the patch antenna resonance
(numERICALLY calculated), \( \tilde{\omega}_{\text{ISB}} \) is the experimental ISB frequency
measured in a multi-pass waveguide configuration (it includes, therefore,
the plasma polarization shift and, hence, the tilde sign), \( \Gamma_{\text{opt}} \) is the
electromagnetic overlap factor with the active region, \( f_w \) is the ratio
between the QW width and the semiconductor active region thickness, and $\omega_{\text{plasma}}$ is the ISB plasma frequency (obtained from the sample doping). The curve was fitted to the far-field data by adjusting $E_{\text{opt}}$. Strong coupling theory clearly explains both far-field and near-field spectra, at odds with a zero-coupling simulation of the antenna resonance frequency vs $L$ shown as a dashed green line.

Substantial agreement between far-field and near-field spectroscopy data is observed, with the possible exception of the upper polariton branch for $L < 1.7 \mu m$, where the upper peak is blue-shifted compared to the far-field data and to the theory. This is confirmed by numerical simulations (Comsol) of the local electric field intensity, calculated along the patch side on which the radiation impinges (map in the background). The EPON resist refractive index was adjusted to $n = 1.87$ to fit the observed Rabi splitting energy. We also verified that this combination would lead to the same overlap factor for the secular equation. The simulation can be taken as an estimate of the near-field scattering efficiency measured in SNOM and is also blue-shifted compared to the green curve and blue dots in the upper polariton branch for $L < 1.7 \mu m$. Further studies are required to understand the small discrepancy observed between near-field and far-field resonances, but the general picture emerging from this work confirms that near-field observation of strong light-matter coupling in individual resonators yields consistent results. The SNOM spectra taken on single antennas show that there is no key difference in far-field and near-field analysis of strong-coupling systems and, therefore, that near-field spectroscopy analysis of single resonators with small resonator modal volume $V$ is possible. For example, near-field spectroscopy probes could be used in future photonic integrated chips that would contain single resonators but certainly not entire arrays.

In conclusion, we have realized a series of patch-antenna cavities with varying resonance frequencies in the mid-infrared. The cavities contain a doped QW, whose optical transition is strongly coupled—despite the presence of a relatively thick layer of absorbing dielectric material—to the antenna resonance for specific values of the patch size. We have studied the far-field extinction spectra of arrays and the near-field scattering spectra of single antennas. Both sets of spectra revealed the two polariton branches emerging from operation in the strong light-matter coupling regime. Numerical simulations and analytical model predictions for strong coupling were verified in both experiments, indicating that strong-coupling features are intrinsic to single antenna cavities of subwavelength dimensions. The results are relevant for future nanoscale quantum devices exploiting patch cavities, such as non-linear meta-surfaces or efficient, fast-response mid-infrared detectors.

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### DATA AVAILABILITY

Meaningful data are provided in the text and the rest are available upon request by any reviewer.

### REFERENCES


