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Tracing terahertz plasmon polaritons with a tunable-by-design dispersion in topological insulator metaelements

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Abstract

Collective oscillations of massless charge carriers in two-dimensional materials—Dirac plasmon polaritons (DPPs)—are of paramount importance for engineering nanophotonic devices with tunable optical response. However, tailoring the optical properties of DPPs in a nanomaterial is a very challenging task, particularly at terahertz (THz) frequencies, where the DPP momentum is more than one order of magnitude larger than that of the free-space photons, and DPP attenuation is high. Here, we conceive and demonstrate a strategy to tune the DPP dispersion in topological insulator metamaterials. We engineer laterally coupled linear metaelements, fabricated from epitaxial Bi₂Se₃, with selected coupling distances with the purpose to tune their wavevector, by geometry. We launch and directly map the propagation of DPPs confined within coupled meta-atoms via phase-sensitive scattering-type scanning near-field nanoscopy. We demonstrate that the DPP wavelength can be tuned by varying the metaelements coupling distance, resulting in up to a 20% increase of the polariton wavevector $\text{Re}(k_p)$ in dimers and triplets with a 1 μm spacing, with reduced losses and a >50% increase of the polariton attenuation length.

Introduction

Surface plasmon polaritons (SPPs)¹—hybrid light–matter modes involving collective oscillations of mobile charges that propagate along material’s surface²—can enable extreme light localization, hence opening interesting prospects in optics, microscopy, sensing, and Raman spectroscopy³. They can also modify photon emission rates and be exploited for the development of nanoscale lasers⁴, thanks to the combination of mode confinement to deeply sub-wavelength length scales (down to <1/1000 the free-space wavelength) and enhanced electric field strengths. SPPs have recently been exploited in sub-wavelength plasmonic nanostructures and resonators capable of manipulating light at the nanoscale⁵, an approach particularly effective at far-

infrared wavelengths (>60 μm), where plasmonic nanostructures can help circumvent severe limitations imposed by diffraction, enabling sub-diffraction-limit light–matter interaction and enhanced nonlinear phenomena⁶.

Far-infrared sub-wavelength metallic resonators have been implemented as individual elements and as designer metasurfaces for frequency filtering⁷, wavefront shaping⁸, beam steering⁹, polarization rotation^{10,11} and ultra-strong light–matter coupling¹², albeit without being tuneable. In contrast, scalable two-dimensional (2D) materials¹³, which support deeply sub-wavelength surface plasmon-polariton quasiparticles^{2,14}, offer electrical and optical tunability¹⁵.

Mapping far-infrared SPPs or Dirac plasmon polaritons (DPPs) in 2D materials has become a prominent research area in recent years^{16–25}. At THz frequencies, the polariton dynamics is strongly influenced by the properties and density of charge carriers due to intraband absorption (free-carrier response). Furthermore, Dirac materials, such as graphene, Weyl semimetals, and topological insulators (TIs), exhibit low electronic specific heat

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because of the reduced density of states near the Dirac point²⁶. As a result, THz excitation can lead to huge thermal gradients (up to ~1000 K in graphene²⁷), leading to non-equilibrium distribution of particles within the band structure with a significant change in the optical conductivity. The latter, in turn, results in an intensity-dependent optical response, i.e., strong optical non-linearity^{27,28}.

Topological insulators, such as Bi₂Se₃, support THz DPPs at the surface^{18,20,24,29–31} with high momentum, which can be modified via plasmon coupling to phonons and massive charge carriers in the bulk and at the surface³². In principle, TIs allow tailoring DPP dispersion properties through proper resonator designs and/or lateral coupling between a set of neighboring TI resonators. Recently, a powerful methodology for investigating DPPs in mechanically cleaved¹⁷ and large-area Bi₂Se₃ samples grown by molecular beam epitaxy (MBE)^{18,23} has been developed using scattering-type scanning near-field optical microscopy (s-SNOM)³³. More recently, it was applied to detect and characterize hybrid polaritons in Bi₂Se₃ antennas, which proved to be effective in confining the propagation of DPPs to a single dimension, thereby enhancing the visibility of DPPs despite the strong intrinsic attenuation²⁴.

Here, by using THz-s-SNOM with laser feedback interferometry^{34,35} in THz quantum cascade lasers (QCLs), we reveal a strong effect of inter-resonator coupling in Bi₂Se₃ doublet or triplet antennas, and show that the DPP dispersion can be modified by controlling the lateral gap between lithographically defined antennas. We determine dispersion properties of coupled DPPs propagating in the antennas at THz frequencies and show that the inter-resonator coupling leads to an increase of the DPP momentum by over 20%, resulting in values more than one order of magnitude higher than the free-space photon momentum. While the real part of the momentum increases, the loss remains similar to the case of uncoupled resonators, suggesting that the inter-resonator coupling strategy can mitigate the effects of strong DPP intrinsic losses observed in 2D materials³⁶. By carefully choosing coupling distances and resonator design, one could enable quantum devices exploiting the properties of DPPs. This result has significant implications for the development of integrated plasmonic devices that rely on sub-wavelength metaelements for manipulating THz radiation. Furthermore, it holds promise for applications in quantum information³⁷, spintronics³⁸, THz photonics³⁹, and laser physics⁴⁰.

Results and discussion

The THz s-SNOM apparatus, adopted in the present work, operates in a phase-sensitive detector-less configuration through self-mixing (SM) interferometry, with a

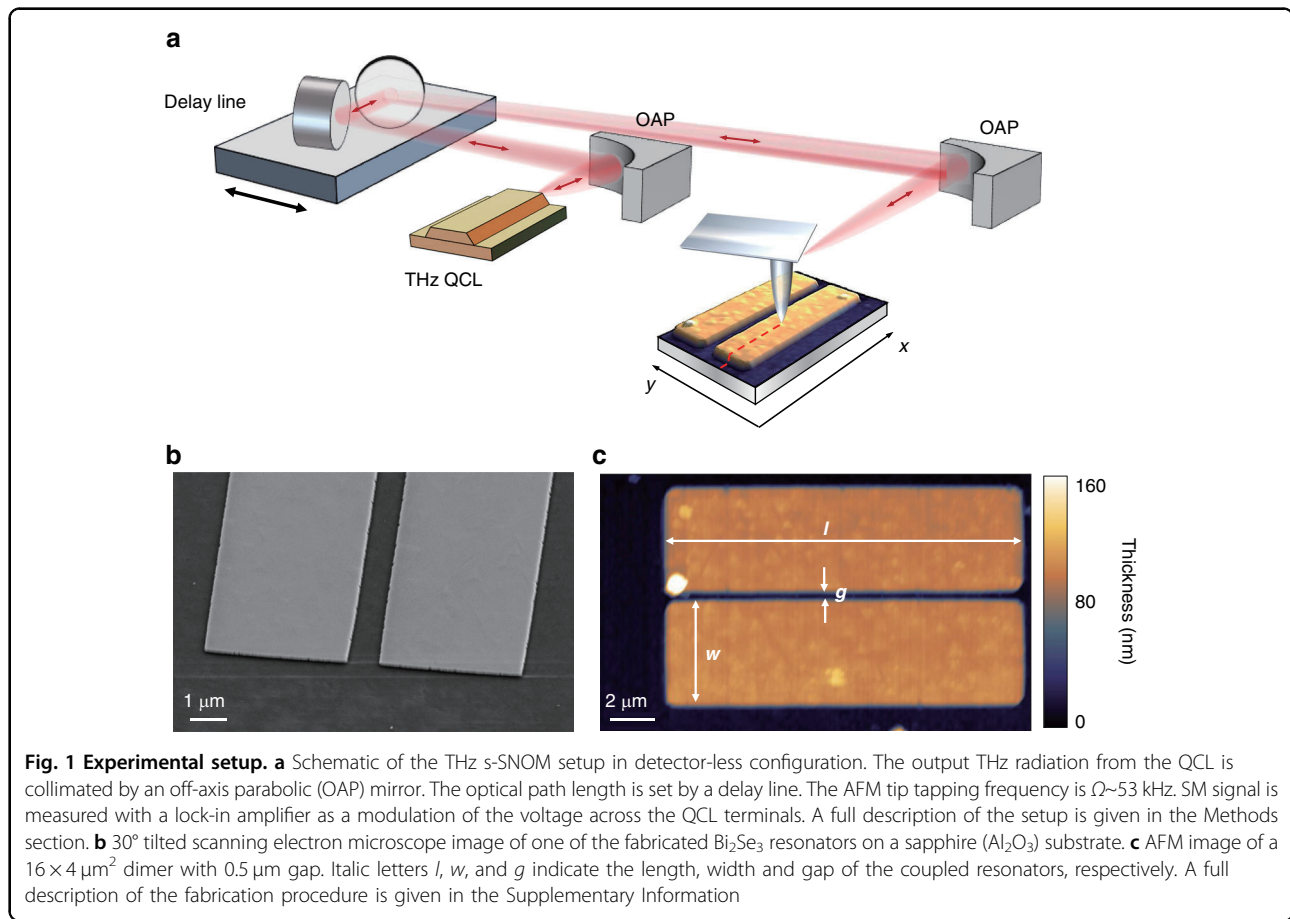
set of three THz QCLs^{34,35}. This method has proven instrumental for launching and capturing the propagation of THz plasmon polaritons in samples with reduced dimensionality, via the real-space mapping of surface waves with wavelengths smaller than $\lambda_0/100$ ^{21,41}, where λ_0 is the wavelength of the free-space propagating wave. The laser beam is focused on an atomic force microscope (AFM) tip, which acts as a local plasmon excitation source when positioned above the sample surface. Figure 1a shows a schematic of the experimental setup (see Supplementary Information for further details). The illuminated tip induces a strongly confined polarization in the surface, and thus it can couple the incident beam to a surface-guided polariton with a wavevector much higher than that of the free-space propagating wave. In our Bi₂Se₃ resonators, the polariton travels along the antenna, reflects off the antenna ends and returns towards the AFM tip, which then scatters it back to the QCL. The scattered wave couples into the laser cavity and mixes with the field inside the cavity, resulting in a change of the QCL terminal voltage. For a tip tapping near the sample surface (~200 nm distance) with a frequency Ω , the terminal voltage therefore displays oscillation at the fundamental frequency and its higher harmonics ($n\Omega$), enabling the detection of extremely weak scattered waves, without the need for a THz detector.

This modulation of the QCL voltage, i.e., the near-field SM signal, carries information about the amplitude and phase of the launched DPPs, detectable even at the 5th harmonic frequency³⁵. The phase is extracted readily by varying the optical path length L between the QCL and the AFM tip, by means of an optical delay line, and recoding an interferometric pattern. The adopted SM detection scheme is inherently fast due to the picosecond lifetime of carriers in QCLs⁴². Therefore, by varying L with a constant velocity during the acquisition of a two-dimensional (x, y) s-SNOM image, it is possible to reconstruct an optical hologram $H_n(x, y) = A_n(x, y) \times \exp(ip_n(x, y))$, enabling phase (ϕ_n) and amplitude (s_n) 2D near-field imaging at the n th order harmonic $\tilde{\sigma}_n(x, y) = s_n(x, y) \times \exp(i\phi_n(x, y))$ by means of post-processing⁴³. Here, we focus on phase detection for accurate measurement of the dispersion of plasmon-polaritons with extremely short propagation lengths²⁴. We concentrate our analysis on the third demodulation order, which is chosen as a good compromise between signal intensity and far-field background suppression^{34,35}.

The adopted QCL-based holographic approach⁴³ enables quantitative retrieval of the plasmon phase, eliminating the need for external photodetectors and interferometric homodyne⁴⁴, heterodyne⁴⁵ or pseudo-heterodyne^{46,47} schemes.

The dispersion of DPPs is influenced by the mobility and the density of electrons in the topological surface states in Bi₂Se₃^{18,32}. In the THz spectral region, it is also

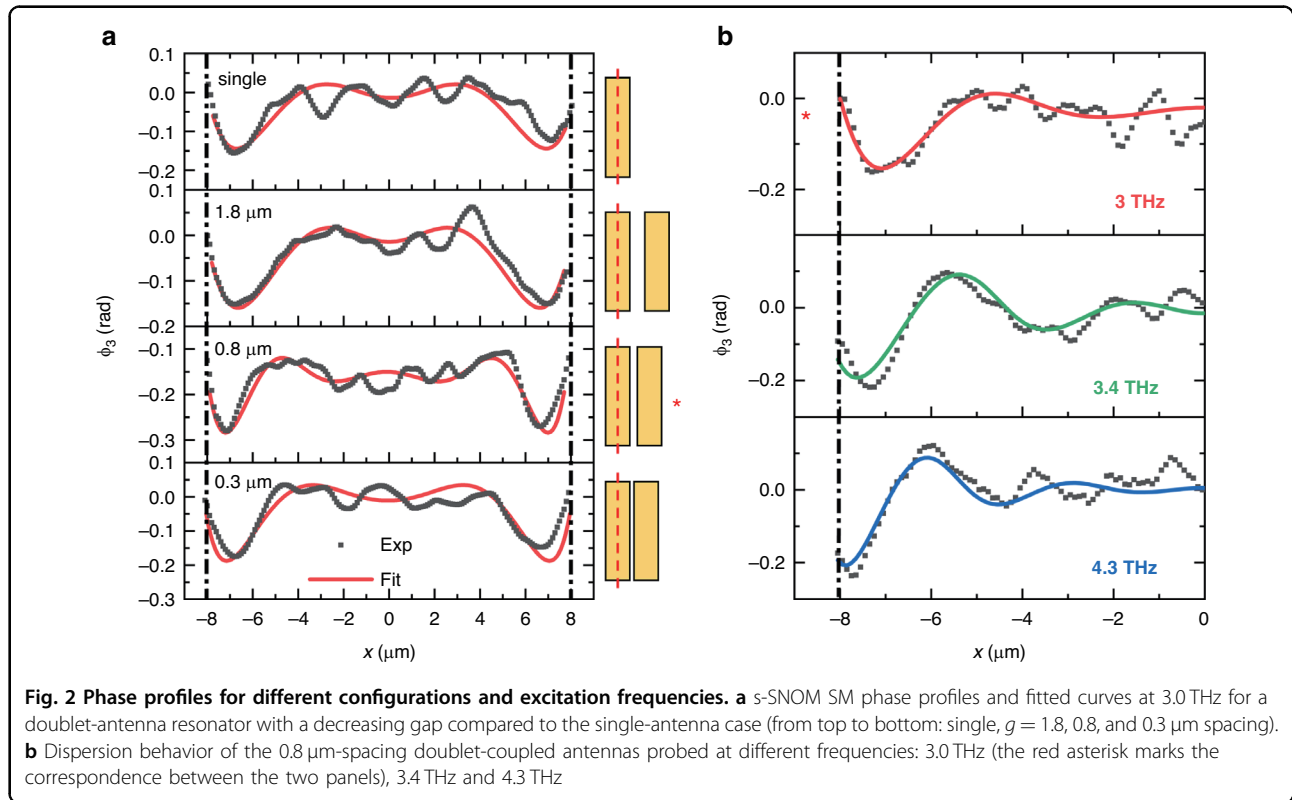




affected by two infrared active phonon modes at 1.92 THz and 4.05 THz³², as well as by free massive carriers in the surface and bulk states^{18,23}. THz DPPs in Bi_2Se_3 have been thoroughly characterized using near-field real-space mapping of plasmons on MBE-grown large-area layers^{18,23} as well as lithographically defined antenna resonators²⁴. We selected three single-mode QCLs emitting in the spectral range near the two phonon resonances: at 3.0 THz, 3.4 THz, and 4.3 THz. First, we characterized and verified the dispersion of surface plasmons in a single antenna^{18,23,24}, and then we compared the k -vector in a doublet and a triplet antenna with the reference single-antenna case.

Our rectangular antennas were designed to be approximately twice as long as the expected DPPs wavelength at 3 THz. For this antenna length, tip-launched DPPs produce at least two full oscillations of SM phase signal ϕ_n along the antenna²⁴, sufficient for determining the k -vector. We fabricated antennas that are $16 \mu\text{m}$ long (l) and $4 \mu\text{m}$ wide (w). These were etched from an 80 nm thick continuous film of MBE-grown Bi_2Se_3 on sapphire (details about MBE growth are provided in the Supplementary Information). Scanning electron microscope (SEM) images and AFM topography of the fabricated

coupled resonators are shown in Fig. 1b, c (details about the fabrication procedure are provided in the Supplementary Information). The rectangular antenna geometry, with one dimension much shorter than the other, allows us to confine the polariton propagation to a single dimension, avoiding the geometrical decay⁴⁸ caused by the radial divergence of tip-launched waves. The top panel in Fig. 2a shows the profile of ϕ_3 along a single antenna, where the phase clearly displays an oscillatory behavior starting at the edges and decaying toward the antenna center. The spatial frequency of these oscillations and their decay allowed us to determine real and imaginary parts of the k -vector of the tip-launched plasmon by fitting the model of plasmon propagation along the antenna to the experimental data (see Supplementary Information). The results for the single antenna are consistent with previously reported values²⁴. By comparing the profile shown in Fig. 2a for a Bi_2Se_3 resonator with that obtained from a gold resonator of identical shape (see Supporting Information), we can unveil the presence of structured noise in our experiments. To study how the SPP spatial frequency changes when another antenna of the same geometry is placed near and parallel to the first antenna, we varied the gap (g) between antennas in two



geometries—in the doublet and triplet antennas. We found that the size of the gap significantly affects the observed phase profile.

Figure 2a shows the results obtained for three different values of doublet gap (g), 1.8, 0.8, and $0.3 \mu\text{m}$, indicating varying optical coupling. For the $0.3 \mu\text{m}$ gap in the doublet geometry, the spatial frequency decreased slightly compared to the single isolated antenna case; the spatial frequency remained similar to that retrieved in the single antenna for the $1.8 \mu\text{m}$ gap. Conversely, for the $0.8 \mu\text{m}$ gap, the spatial frequency visibly increased. These changes in ϕ_3 profiles along the antenna axis indicate that the plasmon dynamics in the doublet and triplet antenna resonators are strongly influenced by the lateral near-field coupling.

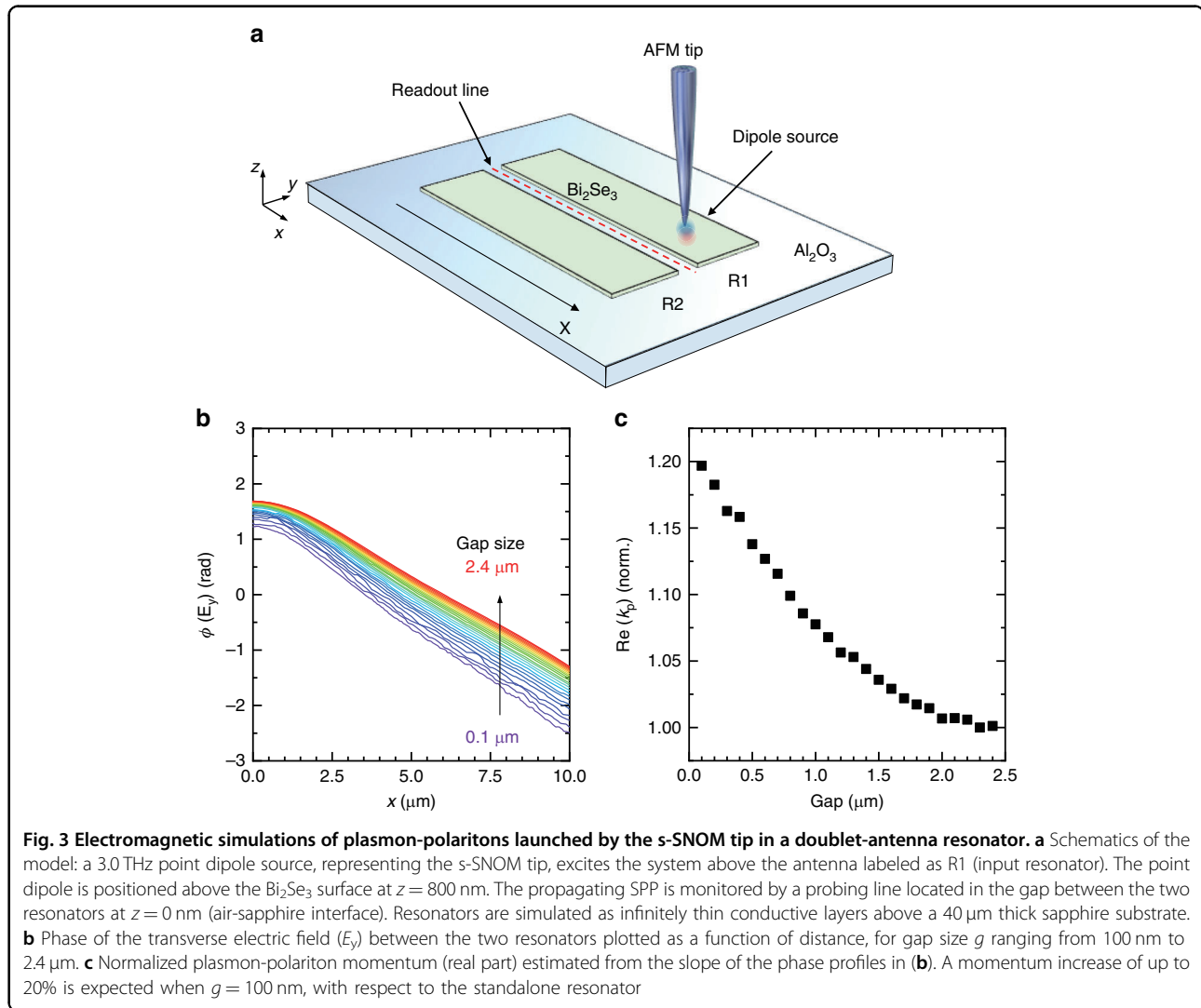
We also considered the case of $0.8 \mu\text{m}$ gap and used different QCLs to excite plasmons at higher frequencies. Figure 2b shows the ϕ_3 profiles acquired along the doublet resonator for excitation frequencies of 3.0, 3.4, and 4.3 THz. In all cases, the plasmon-polariton k -vector (k_p) increased in the doublet compared to the single-antenna case. Indeed, the corresponding wavelength is reduced in the doublets, as evidenced by the comparison between the red fitting curves in Fig. 2a.

To gain an understanding of the antenna gap effect, we conducted electromagnetic simulations of plasmon coupling between two long adjacent antennas in the doublet resonator. Electromagnetic simulations were performed

using a commercial software based on a finite element method (Comsol Multiphysics^{®49}, see “Methods” section for further details). In the simulated model (Fig. 3a), one resonator (R1) was excited by a dipole point source, mimicking the excitation produced by the s-SNOM tip. We then observed the propagation of the DPP along the antenna axis. To ensure the DPP propagation over a sufficiently long distance, we modeled the antenna to be $30 \mu\text{m}$ long, i.e., equal to several plasmon attenuation lengths. By analyzing the in-plane component of the electric field (E_y) in the gap between the antennas, we extracted the plasmon k -vector. As the gap (g) between the two resonators is decreased from $2.4 \mu\text{m}$ to 100 nm , we observed that the rate at which the phase of E_y changes progressively increases (Fig. 3b). This suggests that the k -vector increases as the gap becomes smaller. Figure 3c illustrates the relationship between the real part of the wavevector $\text{Re}(k_p)$ and g , showing a progressive decrease in $\text{Re}(k_p)$ as the resonators are moved further apart.

From the analysis of the phase oscillations in Fig. 2 we obtain the complex value of the plasmon-polariton wavevector $k_p = \text{Re}(k_p) + i\text{Im}(k_p)$, which encodes the polariton propagation characteristics, in particular the polariton wavelength $\lambda_p = 2\pi/\text{Re}(k_p)$ and its damping length $L_p = 1/\text{Im}(k_p)$. The detailed fitting procedure is described in the Supplementary Information. The results of the analysis are shown in Fig. 4, which displays the extracted dispersion (Fig. 4a–c) and damping (Fig. 4d–f) retrieved for all the





measured coupled resonator configurations, at the three frequencies under study: 3.0, 3.4, and 4.3 THz, indicated as dashed horizontal lines (Fig. 4b, c). Figure 4a displays the measured DPP dispersion for a standalone $16 \times 4\text{ }\mu\text{m}$ resonator, demonstrating a wavelength compression ratio $(\lambda_0/\lambda_p) \sim 10$, consistent with previous reports¹⁸. The experimental data are compared to the polariton dispersion predicted by an analytical conductivity model^{18,21,50}. In this model, the Bi_2Se_3 layer is considered as a layer of zero thickness, with a total conductivity (σ_t) given by the sum of bulk (σ_{bulk}), massive surface carriers ($\sigma_{2\text{DEG}}$), and Dirac carriers contributions (σ_{DC}). The complex polariton wavevector k_p is evaluated as $k_p = ik_0\epsilon_r \times (c/2\pi\sigma_t)$ ¹⁸, where k_0 is the free-space wavevector, $\epsilon_r = (\epsilon_{\text{sub}} + \epsilon_{\text{sup}})/2$ is the average permittivity of the materials surrounding the Bi_2Se_3 layer (substrate and superstrate), and c is the speed of light. The best agreement between the experimental data points and the analytical model is achieved with the following values of carrier densities: $n_{\text{bulk}} = 1.8 \times 10^{19}\text{ cm}^{-3}$, $n_{\text{DC}} = 1.0 \times 10^{13}\text{ cm}^{-2}$, and

$n_{2\text{DEG}} = 0.4 \times 10^{13}\text{ cm}^{-2}$, in good agreement with previous reports on similar structures²⁴. Further details on the analytical model and on the evaluation of the three conductivities, σ_{bulk} , $\sigma_{2\text{DEG}}$, and σ_{DC} , are provided in the Supplementary Information.

We then investigated how the dispersion of DPPs is modified when the Bi_2Se_3 antennas are laterally coupled in dimers or triplets. Figures 4b, c display the obtained results, indicating that the DPP's dispersion is notably different when the resonators are separated by a gap $g = 0.8\text{ }\mu\text{m}$. Specifically, the polariton wavevector $\text{Re}(k_p)$ is increased by over 20% in dimers and triplets with this gap, compared to the predicted trend (dashed lines in Figs. 4b, c) for the case of standalone resonators. Interestingly, the coupling strength η , quantified here as the percentage increase of $\text{Re}(k_p)$ relative to the standalone resonator, exhibits a non-monotonic dependence on the gap g between the two coupled resonators: η has a maximum value for $g = 0.8\text{ }\mu\text{m}$. We attribute this observation to the



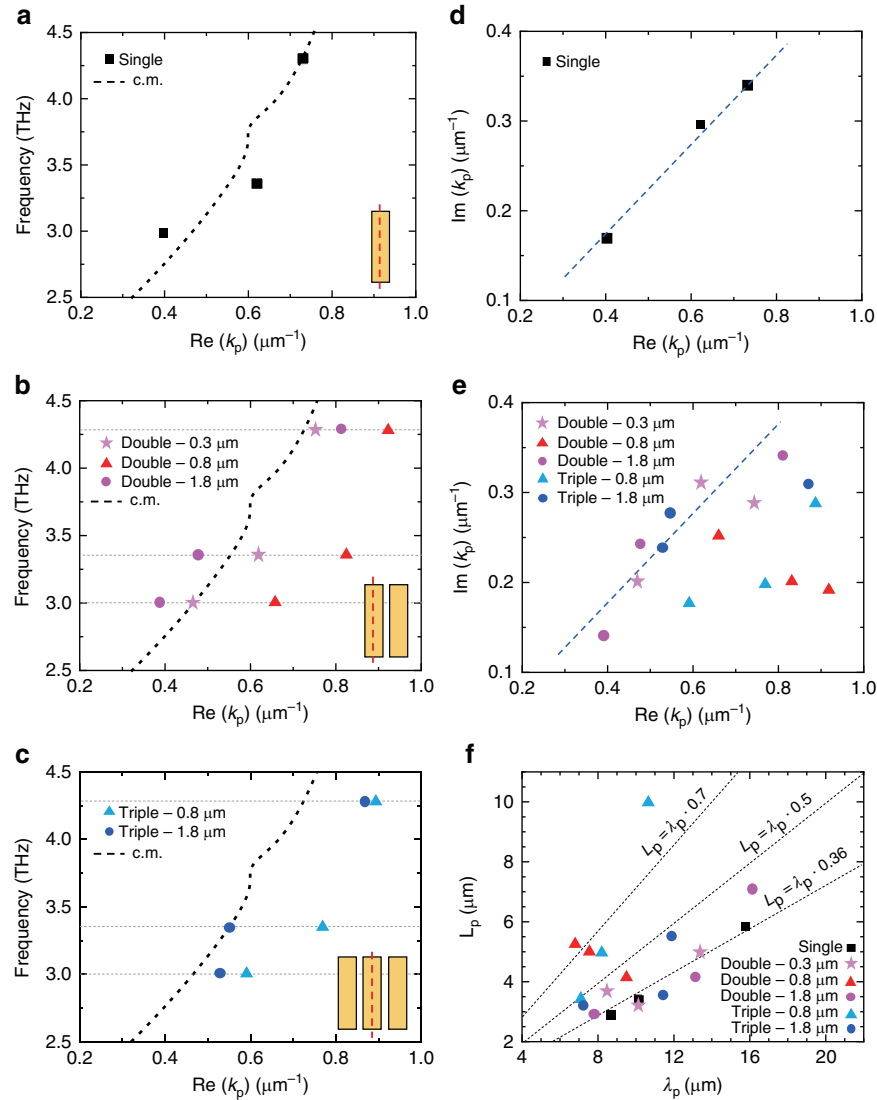


Fig. 4 Polariton dispersion for different device configurations. **a** Polariton dispersion obtained by fitting the experimental phase profiles for the single antenna. The black dashed line corresponds to the SPP dispersion predicted by an analytical conductivity model (c.m.), described in the Supplementary Information. **b, c** Polariton dispersion for the doublet (**b**) and triplet (**c**) configurations. The black dashed lines correspond to the lines in (**a**), facilitating a direct comparison with the single-antenna case. Horizontal lines indicate the three investigated frequencies: 3.0, 3.4, and 4.3 THz. **d** Imaginary part vs. real part of the polariton wavevector for a standalone resonator. The blue dashed line is a guide to the eye. **e** Imaginary part as a function of the real part of the polariton wavevector in the cases of doublets and triplets. Dashed lines correspond to those in (**d**). **f** Polariton decay length (L_p) as a function of the polariton wavelength (λ_p). The dashed lines represent different proportional relationships between the two quantities. The line $L_p = \lambda_p \times 0.36$ is derived from a fit to the single-resonator measurements

fact that, for $g = 0.3 \mu\text{m}$, the separation between two neighboring antennas is small enough that the SPP is transmitted through the gap with negligible reflection.

The effect of lateral coupling can also be observed in the analysis of the DPP propagation length L_p . Figure 4d presents the $\text{Im}(k_p)$ values plotted against $\text{Re}(k_p)$ for all QCL frequencies for the standalone resonator. In this configuration, the ratio $\text{Re}(k_p)/(2\pi \times \text{Im}(k_p)) = L_p/\lambda_p$, which represents the relative attenuation length of the SPP, remains almost constant regardless of frequency at approximately 0.36.

This value is consistent with that reported in previous studies on MBE-grown Bi_2Se_3 thin films²⁴ and can be attributed to a smaller wavelength compression ratio λ_0/λ_p compared to other material systems, such as hBN-encapsulated graphene. Interestingly, the relative attenuation length is significantly improved in efficiently coupled resonators, as shown in Fig. 4e, which plots $\text{Im}(k_p)$ as a function of $\text{Re}(k_p)$ for various configurations of coupled resonators. In this figure, data points to the right of the diagonal dashed line represent configurations with



lower attenuation. This observation can be further clarified by visualizing the same results in the (L_p, λ_p) plane (see Fig. 4f), where we observe that for a gap spacing of $g = 0.8 \mu\text{m}$ and frequencies above 3.4 THz, the relative attenuation length L_p/λ_p is approximately doubled with respect to that of the standalone antenna, indicating that the propagation losses are strongly reduced in laterally coupled systems. This loss reduction can be intuitively understood as a result of decreased lateral confinement of the propagating SPP along the antenna, due to the evanescent coupling with neighboring structures. However, when the confinement is further reduced ($g = 0.3 \mu\text{m}$), the surface mode can spread across the separation gap, resulting in standard geometrical damping and canceling out the benefits of the *quasi*-one-dimensional propagation. For larger gap spacing ($g = 1.8 \mu\text{m}$), the coupling between neighboring resonators is strongly reduced, leading to propagation losses similar to those observed for the standalone antenna.

Conclusions

In this study, we demonstrate that the wavevector of plasmon polaritons on the surface of Bi_2Se_3 resonators can be tuned by selecting the gap between laterally coupled resonators. These have been designed and fabricated from large-area MBE-grown Bi_2Se_3 thin films. The propagation of plasmon polaritons on the resonator surface has been characterized using THz detector-less s-SNOM, enabled by laser feedback interferometry in QCLs. Near-field measurements allow us to estimate the complex polariton wavevector at 3.0, 3.4, and 4.3 THz. The investigation of tip-launched SPPs propagating on the TI surface shows that the gap (g) between adjacent resonators plays a crucial role in determining the polariton dispersion and dynamics. Importantly, g not only affects the wavelength but also influences the damping losses of the SPP modes excited on the TI surface. Our results demonstrate that it is possible to customize the spectral response of Bi_2Se_3 -based THz resonators by adjusting the gap. This knowledge can be adopted as a design strategy for the implementation of TI-based architectures, taking advantage of various effects such as resonance line narrowing, resonance mode-splitting, and cross-polarization conversion, typically observed in coupled resonator systems. Additionally, we envision that lithographically designed THz metasurfaces using large-area TI materials with specific spectral responses could have a significant impact on far-infrared optoelectronics and nonlinear nanophotonics.

Materials and methods

Electromagnetic simulations

Electromagnetic simulations were performed using commercial software (COMSOL Multiphysics®), based on

a finite element method. The resonators in the simulation geometry are long enough to contain multiple plasmon polaritons wavelengths and to be much longer than the polaritons damping length ($30 \times 4 \mu\text{m}$). They are modeled as infinitely thin conductive surfaces⁵⁰ located on the substrate (xy) plane, with sheet conductivity σ_{2D} ranging from 10^4 to 10^5 S m^{-1} . We observed some variation in the results using different sheet conductivity values σ_{2D} (data in Fig. 3b are obtained for $\sigma_{2D} = 10^5 \text{ S m}^{-1}$). The sapphire substrate was $40 \mu\text{m}$ thick, with dielectric constant $\epsilon_{\text{Al}_2\text{O}_3} = 10.4$, with the surface plane defined as $z = 0 \text{ nm}$. Surface plasmon in the electromagnetic simulations were generated by a point dipole source (frequency 3.0 THz) aligned along the out-of-plane direction located $z = 800 \text{ nm}$ above the resonator surface (input resonator, R1). The propagation of the SPP was analyzed by extracting the transverse component of the electric field E_y at $z = 0 \text{ nm}$ in xy -plane. In the case of Fig. 3, we monitor the distribution of E_y along the x -axis at the center of the gap between the two resonators.

Acknowledgements

This work was supported by the European Research Council through the ERC project TeraScan (101157731), by the European Union under the Italian National Recovery and Resilience Plan (NRRP) of Next Generation EU, partnership on PE0000023-NQSTI, by the second half of the 2016 Balzan Prize for Applied Photonics delivered to Federico Capasso and the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering. This article describes objective technical results and analysis. The views expressed in the article do not necessarily represent the views of the U.S. DOE or the United States Government. Z.W. and S.L. acknowledge funding from the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Award DE-SC0017801.

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Author contributions

O.M. and M.S.V. conceived the experiment. L.V. fabricated the devices, performed numerical simulations, and analyzed the data. C.S. and L.S. performed the experiments and analyzed the data. Z.W. and S.L. grew the material. The manuscript was written by L.V. and M.S.V. M.S.V. supervised the work. All authors contributed to the final version of the manuscript and discussed the results.

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

Code availability

The codes and simulation files that support the plots and data analysis within this paper are available from the corresponding author upon reasonable request.

Conflict of interest

Miriam Serena Vitiello serves as a Guest Editor of Special Issue on Advances in Science of Light: Research Highlights in Italy for the Journal. No other author has reported any competing interests.



Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41377-025-01884-0>.

Received: 14 October 2024 Revised: 13 April 2025 Accepted: 4 May 2025
Published online: 26 August 2025

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