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Twist-engineered acoustic plasmon nanocavities enable deep-nanoscale terahertz molecular fingerprinting

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Abstract

Field-enhanced terahertz spectroscopy serves as a powerful analytical tool for biochemical sensing, materials characterization, and medical diagnostics, where detection sensitivity fundamentally depends on electric field enhancement and confinement. However, the severe scale mismatch between long terahertz wavelengths (on the order of 100 μm) and nanoscale analytes (typically < 10 nm) imposes critical limitations on conventional far-field techniques. Here, we present a breakthrough sensing approach utilizing twisted double-layer graphene plasmonic metasurfaces (t-DL-GPMs) with nanometric dielectric spacers. Our investigations reveal that these t-DL-GPMs support strongly confined acoustic plasmon nanocavity modes featuring extraordinary field enhancement and deep subwavelength field concentration, along with an extremely small mode volume ($10^{-13}\lambda_0^3$). Compared to single-layer and untwisted double-layer graphene configurations, the twisted architecture demonstrates dramatically improved sensing performance, with figures of merit enhanced by factors of 22 and 48, respectively. Most significantly, the platform enables clear identification of terahertz vibrational fingerprints from molecular layers as thin as 1 nm. This work not only opens novel avenues for nanoscale terahertz detection, pushing the THz sensing into unprecedented deep-nano level, but also establishes a versatile foundation for exploring extreme light-matter interactions and developing advanced terahertz photonic devices.

Keywords: Twisted graphene metasurfaces, Acoustic plasmons, Nanocavity, Terahertz spectroscopy, Sensing

Introduction

Terahertz (THz) field-enhanced molecular spectroscopy has emerged as a powerful analytical tool for material characterization, environmental monitoring, and food safety inspection, owing to its unique capability to provide label-free molecular fingerprint information [1–3]. However, THz far-field spectroscopy suffers from severely limited sensitivity at the nanoscale due to the enormous wavelength mismatch between THz radiation (hundreds of micrometers) and molecular dimensions (typically < 10 nm), leading to a weak interaction between THz waves and the target molecules [4–6]. This

fundamental scaling challenge necessitates extreme field confinement and enhancement to boost light-matter interactions [6–8].

Plasmons—collective oscillations of charge carriers coupled to electromagnetic fields—offer a promising solution through their extraordinary ability to concentrate optical fields into deep subwavelength volumes [7, 9, 10]. Exemplified by metal nanoparticle/nanowire-on-mirror (NPOM/NWOM) plasmonic architectures, these systems exhibit ultra-sensitive plasmonic cavity modes capable of both single-molecule detection and sub-picometer differential resolution [11–14]. While metal-based plasmons have enabled deep nanoscale detection in visible and infrared regimes [11, 15], the performance of the plasmons in metals at THz frequency (often referred to as spoof plasmons) is diminished and lose their optical field confinement capabilities, indicating the ratio of the plasmon wavevector to free-space wavevector (k/k_0) approaches 1, due to the perfect electrical conductor behavior of metals [16]. Although the engineered NPOM-analogous configurations and metal antennas, particularly those with micro-nano scale gaps, can generate strong electric field enhancement and improve the sensitivity of THz spectroscopy [17, 18], the achievable confinement remains insufficient for robust nanomolecular detection, highlighting the need for alternative approaches.

Unlike metals, low-dimensional materials (e.g., graphene, Bi_2Se_3 , Ag_2Te , WTe_2 , black phosphorus) exhibit significantly lower carrier concentrations, red-shifting the plasma frequencies of metals from visible to mid-infrared/THz ranges [16–24]. These materials have emerged as exceptional platforms for THz plasmons due to their inherent nanometer-scale thickness and gate/doping-tunable carrier concentrations [19–22], which enable both strong field confinement and spectral tunability of THz waves. As a result, plasmons in low-dimensional materials hold significant promise for overcoming the challenges of THz nanoscale molecular detections. Graphene plasmons, in particular, have demonstrated remarkable success in mid-IR molecular sensing, detecting 20 nm protein layers via Fabry-Pérot (FP) modes [7]. Notably, graphene plasmons has been demonstrated to offer distinct advantages over metallic plasmons for studying nanoscale molecules [7], primarily due to their superior electric field compression. By further engineering plasmonic cavities within graphene structures, it becomes possible to compress electromagnetic fields into extremely small volume [6, 23, 24]. A notable example is the graphene acoustic plasmon mode that arises in graphene-on-dielectric-metals and double layered graphene configurations [6, 23–27]. This mode has been shown to enhance the sensitivity of mid-IR molecular spectroscopy [6, 22, 24, 28–30]. Nevertheless, to the best of our knowledge, no analogous breakthroughs have yet been achieved in the THz frequencies—a critical knowledge gap that hinders transformative applications in this field.

Recent advances in twistronics have opened new frontiers in nanophotonics by enabling precise control over interlayer plasmonic coupling [31–36]. Compared to graphene-on-dielectric-metals and aligned double layered graphene configurations, the introduction of a twist angle provides a unique degree of freedom to engineer the effective coupling area in twisted graphene systems, possibly enabling the creation of extreme-subwavelength THz nanocavities with exceptional field confinement. This approach could dramatically boost THz-matter interactions, resolving the critical size mismatch between THz wavelengths and target analytes. Despite recent advances in

understanding propagating plasmons in twisted graphene architectures [37], which is unfavorable for strong field enhancement and localization, the fundamental characteristics and applications of localized plasmon modes—crucial for deep-subwavelength nanocavities—remain essentially unexplored [38–40]. Unlocking these phenomena could hold the key to overcoming current limitations in THz molecular spectroscopy.

Here we pioneer a groundbreaking THz sensing platform based on twisted double-layer graphene plasmonic metasurfaces (t-DL-GPMs)—periodic arrays of twisted graphene resonators separated by nanometer dielectric spacers. This architecture theoretically enables THz sensing at the deep-nanometer scale, reaching monolayer molecular sensitivity. Full-wave electromagnetic simulations demonstrate that t-DL-GPMs sustain novel acoustic plasmon cavity (APC) modes, which achieve deep-subwavelength field confinement through hybridization of different ordered FP plasmon modes in the coupled upper and lower graphene resonators. Our analysis reveals that optical properties of the APC mode exhibit strong twist-angle dependence — manifested in tunable absorption spectra, field enhancement, and confinement characteristics — directly attributable to twist-modulated reduction in interlayer coupling area and strength. Crucially, t-DL-GPMs outperform both single-layer graphene and untwisted bilayer structures in refractive index sensing, owing to the formation of APC nanocavities. The engineered nanocavities achieve fingerprint detection for 1 nm-thick biomolecular (e.g., γ -aminobutyric acid, GABA), corresponding to a monolayer molecular sensitivity. This work establishes fundamental design principles for t-DL-GPM-based devices and opens new avenues for on-chip THz technologies, including ultra-sensitive molecular sensors and high-efficiency modulators.

Results and Discussion

Research models and Principle of THz acoustic plasmon nanocavity in t-DL-GPMs

Figure 1 illustrates the schematic design of a THz sensing device tailored for detecting ultrasmall-volume molecules, utilizing a t-DL-GPM. In Fig. 1a, a representative 1D t-DL-GPM based on graphene ribbons is depicted, featuring a specific twist angle of $\theta = 90^\circ$ and separated by a sensing analyte film. To enable dynamic tuning the optical response of the proposed system, electric gating can be adapted to precisely manipulate graphene's Fermi level [7]. Figure 1a shows the tuning scheme for the Fermi level of lower graphene ribbons. This t-DL-GPM-based device can be ideally fabricated by first transferring chemical vapor deposition grown graphene onto a SiO_2/Si substrate using a polymer-assisted wet method. The graphene is then patterned into metasurface structures via electron beam lithography and reactive ion etching before being divided into two arrays. Next, one array is precisely stacked onto the other with a controlled twist angle using polydimethylsiloxane (PDMS) and micro-manipulation alignment techniques. Finally, the back-gate is connected to external voltage sources using flying wires, enabling electrical tuning of the Fermi level [23]. While the fabrication of high-quality, large-area twisted graphene resonators presents technical challenges, ongoing advances in material synthesis are paving the way toward realizing this promising device. To comprehensively analyze the plasmonic properties of the t-DL-GPMs, we theoretically investigated two configurations: a 2D twisted double-layer graphene rectangle array and a 1D twisted graphene ribbon array, as illustrated in Fig. 1c. The detailed geometric parameters including

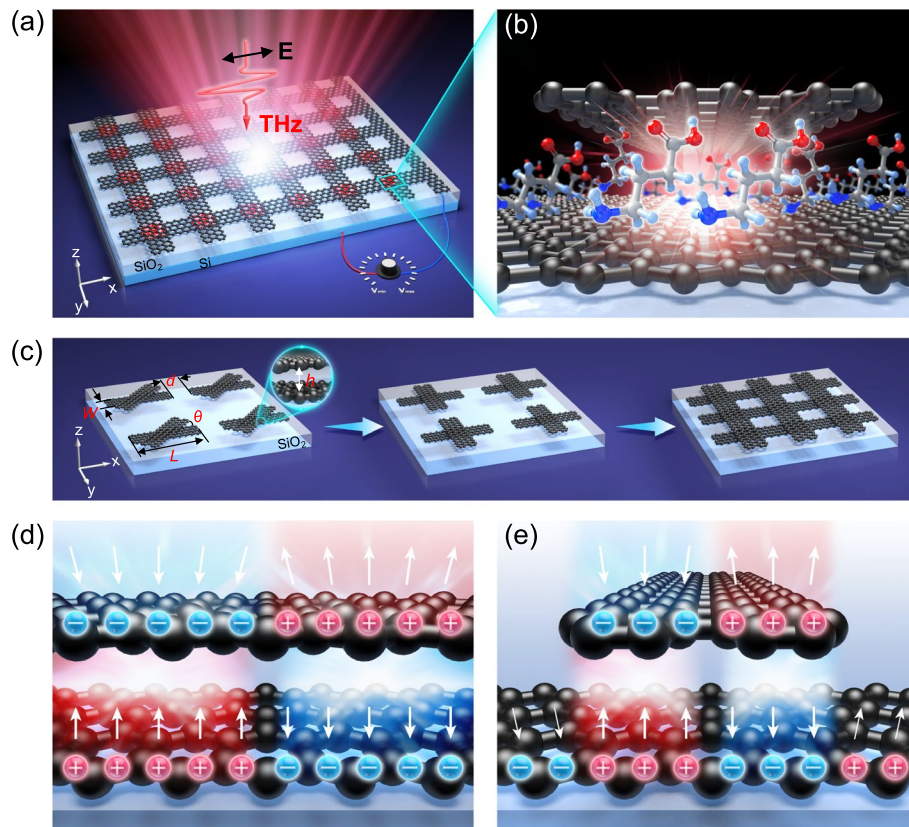


Fig. 1 Concept of a THz sensor based on t-DL-GPMs. **a** 3D schematic of the proposed THz sensor, featuring 1D t-DL-GPM for nanoscale molecular detection. **b** The enlarged view of the spacer in **(a)** where molecules (represented by ball-and-stick model) are adsorbed. **c** Schematics of 2D- and 1D- t-DL-GPMs investigated in this study. Here, d represents the distance between two horizontal resonators, h is the thickness of the spacer between two vertical graphene resonators, W is the width, and L is the length of the graphene resonator. **d**, **e** Schematics illustrating the principle of acoustic plasmon mode under the untwisted case (**d**) and twisted case (**e**). Spheres with “+” and “-” represent charges on graphene layers. The metasurface is illuminated by normally incident THz wave (along the z -axis), with its polarization aligned along the x -axis

twist angle θ , resonator length L , level distance d , width W and vertical distance h are highlighted in left Fig. 1c.

When illuminated by THz waves, graphene plasmon resonators (e.g., ribbons, rectangles) can support a series of localized plasmon modes with ordinal numbers $n=0, 1, \dots$ (corresponding to monopoles, dipoles, etc.) arising from FP resonances mechanisms [41]. These modes exhibit strong dependence on resonator geometry and can be precisely tuned by varying structural parameters [42]. Crucially, in coupled double-layer systems, the Coulomb interaction between phase-matched FP modes in adjacent graphene resonators generates two distinct hybridized plasmon modes: optical and acoustic plasmon modes, the former is characterized by in-phase charge oscillations and the latter is featured out of phase charge oscillations [6, 23, 25–27]. The acoustic plasmon modes are particularly significant for sensing applications due to their ability to strongly confine electric fields within the spacer between the two resonators [6, 23].

The formation of acoustic plasmon modes exhibits fundamentally distinct characteristics in twisted versus untwisted graphene resonator systems. In the aligned configuration

($\theta=0^\circ$), conventional acoustic modes arise through coupling between identical-order FP plasmon modes ($n=n'$) in the vertically stacked resonators, as the schematic shown in Fig. 1d, analogous to graphene-dielectric-metal heterostructures [6, 23]. However, introducing interlayer rotation creates a paradigm shift in plasmon hybridization dynamics. At significant twist angles (e.g., $\theta=90^\circ$), the dramatically reduced effective coupling length between resonators enables unprecedented inter-modal coupling between: (i) different-order FP modes ($n \neq n'$) and (ii) localized FP modes with propagating plasmons, as illustrated by the charge oscillations under the twist case (Fig. 1e). This unique hybridization mechanism generates an ultracompact three-dimensional (3D) acoustic plasmon cavity (APC) that achieves remarkable THz field confinement, compressing THz waves (typically on the scale of hundreds of micrometers) into nanoscale volumes, which is much smaller than the untwisted systems, as the schematics shown in Fig. 1d and e. By adsorbing analyte molecules into the spacer, which can be experimentally achieved by physical adsorption utilizing van der Waals forces or electrostatic interactions, or chemical modification based on non-covalent assembly [7, 12, 14, 43], as shown in Fig. 1b, the intense fields within the 3D nanocavity thus could dramatically enhance the fingerprint signals of the extremely small-volume target molecules, enabling highly sensitive detection.

Plasmon modes analysis of 2D t-DL-GPMs

Let us begin by examining the far-field and near-field plasmonic properties of the rectangle resonators based 2D t-DL-GPM, as illustrated in Fig. 2a. It is important to note that the distance d is 3 μm , which is significantly larger than the decay length of the electric field at the rectangles' edges. As a result, near-field coupling between horizontal resonators does not contribute to the optical properties of the 2D t-DL-GPM, equaling to a single twist graphene rectangle resonators. Figure 2b presents the absorption spectrum (black curve) and electric field spectrum (red curve) of the 2D t-DL-GPM, where four plasmon modes appear at identical positions in both spectra, indicating their strong correlation of far- and near-fields [44]. To elucidate the origins of these modes, we further calculated the electric field (E_z) distributions on the xy -plane (upper panels in Fig. 2c) and the xz -plane (see Supplementary Figure S1) under excitation at the corresponding peak frequencies. The distributions reveal that the electric field is strongly confined within the vertical spacer, a consequence of plasmon coupling between the two vertically stacked resonators. These characteristics suggest that all observed modes are acoustic plasmon modes [6, 22].

In detail, the electric field distribution under 3.3 THz excitation exhibits dipolar characteristics (corresponding to the first-order FP mode, $n=1$) on both the upper and lower resonators, identifying it as the dipolar acoustic plasmon mode (named M_1). In contrast, the field distribution under 4.4 THz excitation shows a dipolar mode ($n=1$) on the upper resonator and a third-order FP mode ($n=3$) on the lower resonator, marked in M_2 . Although the electric field under 7 THz excitation also displays dipolar-like features ($n=1$) on both resonators, the polarization direction is rotated by 90 degrees compared to that at 3.3 THz. This mode is therefore identified as the transverse dipolar acoustic plasmon mode (M_3), while M_1 is referred to as the longitudinal dipolar acoustic plasmon mode. At a higher frequency of 9 THz, the field distribution clearly exhibits a

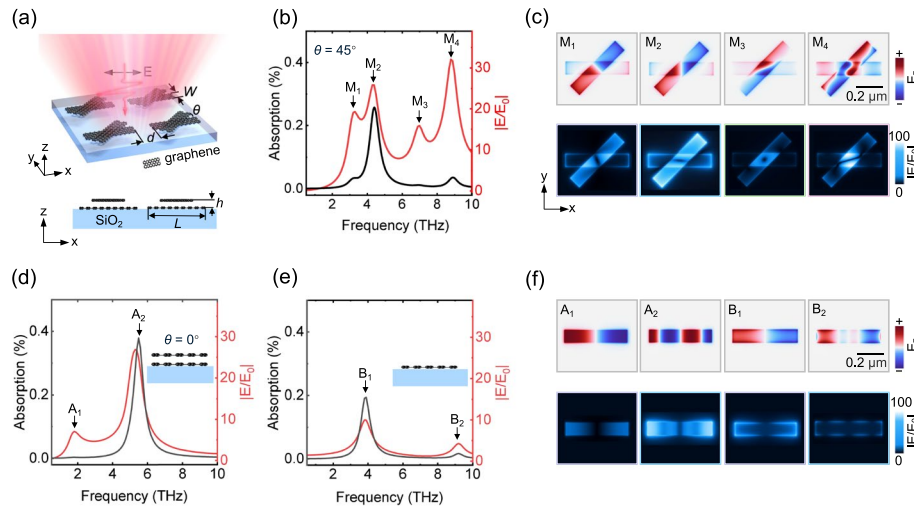


Fig. 2 Plasmon modes analysis in a 2D t-DL-GPM. **a** Schematic of rectangle resonators based 2D t-DL-GPM. Here, d , W , θ , h , and L represent the horizontal distance, the width of the rectangle, the twist angle, the vertical separation between the two rectangles, and the length of the resonators, respectively. **b** Absorption spectrum (black curve) and averaged electric field spectrum (red curve) of the 2D t-DL-GPM. **c** Top panels: E_z field distributions at the peak frequencies indicated in **(b)**. Bottom panels: electric-field enhancement distributions ($|E/E_0|$), where $E_0 = 1 \text{ V/m}$. **d** Absorption (black curve) and averaged electric field spectrum (red curve) of the untwisted 2D DL-GPM. **e** Absorption (black curve) and averaged electric field spectrum (red curve) of the 2D single-layer rectangle metasurface. **f** $|E_z|$ field distributions at the peak frequencies indicated in **(d)** and **(e)**. The average electric field is computed by integrating over the volume of a horizontal rectangular prism (parallel with x -axis) with thickness h , width W and length L . The structural parameters for the rectangle arrays are as follows: $W = 0.1 \mu\text{m}$, $L = 0.5 \mu\text{m}$, $d = 3 \mu\text{m}$, $h = 5 \text{ nm}$, and $\theta = 45^\circ$. The Fermi level (E_F) of graphene is 0.455 eV , and the relaxation time is 0.25 ps . The substrate is SiO_2 . The substrate, E_F and relaxation time of graphene are the same unless we mentioned elsewhere

higher-order mode ($n=3$) on both resonators, classifying it as the third-order acoustic plasmon mode (marked in M_4). Interestingly, by comparing the electric field distributions (lower panels in Fig. 2c), the mode M_4 demonstrates a strong concentration of the electric field within the overlapping region between the upper and lower resonators, providing the highest electric field enhancement (Fig. 2b and c). This results in the formation of a 3D cavity that confines THz waves into a nanoscale volume of $\sim 100 \times 100 \times 5 \text{ nm}^3$, corresponding to as low as $8 \times 10^{-11} \lambda_0^3$ of mode volume (V_m) calculated via a commonly used formula (see Supplementary Figure S2a,b; see Methods for definition). However, this formula neglects graphene's dispersive effects. To address this, we recalculated the mode volume by using a dispersion-corrected formalism (V'_m , see Methods). The revised formalism yield a mode volume of $6 \times 10^{-11} \lambda_0^3$, arising slight reduction in mode volume compared to the non-dispersive case, (see Supplementary Figure S2d,e). Thus, we specifically named this kind of mode as acoustic plasmon cavity (APC) in this work. Furthermore, the portion (η) of energy localized in the interlayer 3D cavity region between the top and lower graphene layers reaches around 73% when normalized to the entire graphene structure for this mode (Supplementary Figure S2f; see Methods for definition), exceeding the corresponding portions from the other modes.

For comparison, we further investigated the plasmonic properties of a 2D untwisted DL-GPM (Fig. 2d) and a 2D single-layer graphene rectangle metasurface (Fig. 2e). In both configurations, two distinct peaks are observed in the absorption and electric field

spectra (marked in A_1 , A_2 and B_1 , B_2 in Fig. 2d and 2e, respectively). For the untwisted DL-GPM, the field distributions (Fig. 2f and Supplementary Figure S1c) reveal that the mode A_1 and A_2 correspond to the first-order ($n=1$) and third-order ($n=3$) acoustic plasmon modes, respectively [6]. In contrast, for the single-layer case, the mode B_1 and B_2 are attributed to the first-order ($n=1$) and third-order ($n=3$) optical plasmon modes [22, 23], respectively. Notably, the electric field enhancements in these two cases, which are primarily localized at the edges of the rectangles, are significantly weaker compared to the APC mode in the 2D t-DL-GPM. This highlights the superior field confinement and enhancement capabilities of APC mode in the twisted configurations.

The stark contrast between the twisted and untwisted configurations underscores the critical role of the twist angle in modulating plasmonic properties. The twisted geometry enables the excitation of APC modes which facilitates the creation of a highly localized and enhanced electric field within the 3D nanocavity. This unique feature makes the 2D t-DL-GPM a promising platform for applications requiring strong light-matter interactions. Further exploration of the twist angle's influence on plasmonic behavior could unlock new opportunities for designing advanced nanophotonic devices.

We further investigated the influence of the twist angle (θ) on the plasmonic properties of the 2D t-DL-GPMs. Figure 3a presents the corresponding contour map of the absorption spectrum as a function of θ . Notably, the APC mode (M_4) emerges at approximately $\theta=5^\circ$ and becomes more pronounced with increasing θ , particularly in the electric field contour map (Fig. 3b). This behavior can be attributed to the interplay between

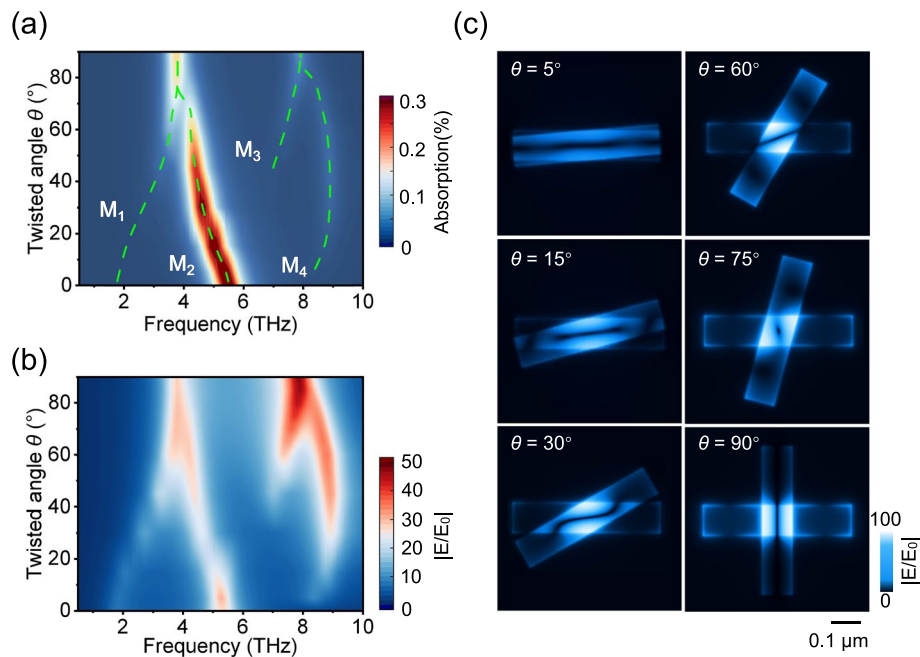


Fig. 3 Plasmonic properties as a function of the twist angle θ in 2D t-DL-GPM. **a** Absorption contour map showing the dependence of absorption on the twist angle θ . The dashed lines serve as visual guides for the eyes. **b** Average electric-field map illustrating the variation of electric field with the twist angle θ . The averaged volume is same as that in Fig. 2b. **c** The electric field enhancement distributions at xy plane for specific twist angles: $\theta = 5^\circ, 15^\circ, 30^\circ, 60^\circ, 75^\circ$, and 90° . The rest structural parameters for the 2D t-DL-GPM are same as that in Fig. 2

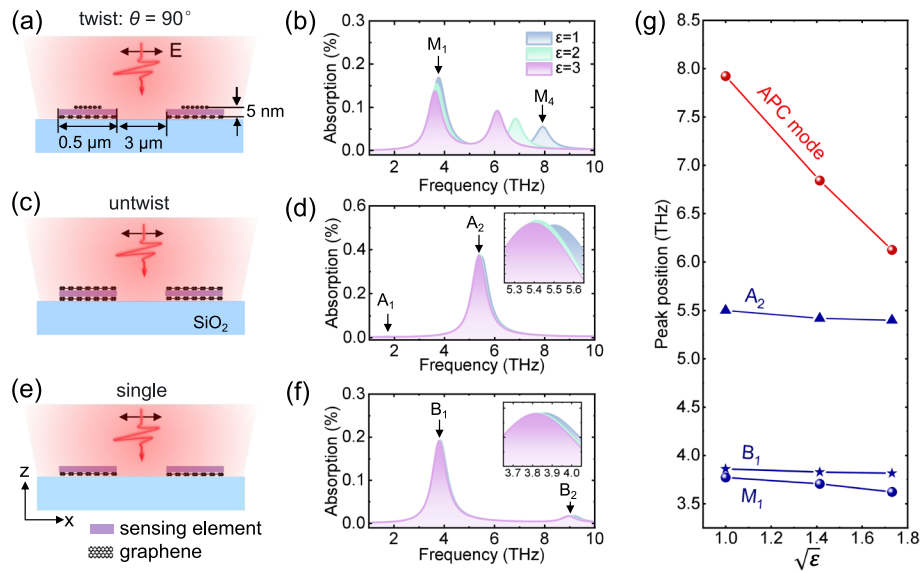


Fig. 4 Performance of THz refractive index sensors based on 2D t-DL-GPM. **a c e** Schematics of sensing based on the 2D t-DL-GPM (**a**), the untwisted DL-GPM (**c**) and the single-layer graphene metasurface (**e**). **b d f** Absorption spectra of the 2D t-DL-GPM (**b**), the untwisted DL-GPM (**d**) and the single-layer graphene resonators (**f**) as ϵ of the sensing element varies from 1 to 3. The sensing elements are shown by purple rectangle in the schematics. The insets show zoomed-in views of A_2 and B_1 , respectively. **g** Dependence of the resonant peak positions on ϵ of the sensing element. The blue-sphere, triangle and star lines correspond to M_1 , A_2 , B_1 in the cases of (**a**), (**c**) and (**e**), respectively. The red-sphere line represents the APC mode in the case of (**a**). The structural parameters for three metasurfaces are as follows: $W = 0.1 \mu\text{m}$, $L = 0.5 \mu\text{m}$, $d = 3 \mu\text{m}$. The vertical spacer in 2D t-DL-GPM is $h = 5 \text{ nm}$

the reduction in the effective coupling area and the enhancement of plasmon coupling intensity due to the formation of a 3D cavity. Specifically, the resonant position of the APC mode exhibits a slight blue shift from 5° to 45° , followed by a red shift. Concurrently, the mode M_1 undergoes a blue shift due to the reduction in coupling length, while the mode M_2 displays a red shift before eventually disappearing, a consequence of the synergy and competition between coupling length and intensity. The mode M_3 appears at around $\theta = 45^\circ$ and vanishes as θ approaches 90° , driven by changes in the coupling strength between the two graphene resonators. Figure 3b illustrates the contour map of the electric field as a function of θ . It is evident that the APC mode (M_4) provides the strongest field enhancement upon excitation. The electric-field enhancement intensifies with increasing θ , reaching its maximum at $\theta = 90^\circ$. This observation is further corroborated by the electric-field distributions under the excitation of the APC mode (M_4), as shown in Fig. 3c.

THz refractive index sensor with the 2D L-DL-GPMs

The ultra-high field confinement and enhancement of the APC mode in the 2D t-DL-GPM make it highly suitable for sensing trace amounts of analytes placed within the spacer. To evaluate the sensing capability of the APC mode, we simulated its spectral shifts as a function of the dielectric permittivity (ϵ) of the spacer. Figure 4b illustrates the spectral shifts of the APC mode (M_4 in Fig. 2b) and the dipolar acoustic plasmon mode (M_1 in Fig. 2b) for the 2D t-DL-GPM with $\theta = 90^\circ$ (schematic shown in Fig. 4a).

For the APC mode (M_4), the resonant peak shifts by $\Delta f = 1.8$ THz as ε changes from 1 to 3 (the red line in Fig. 4g). In contrast, the M_1 mode exhibits a smaller shift of $\Delta f = 0.2$ THz (the blue-sphere line in Fig. 4g). For comparison, we performed analogous calculations for the 2D untwisted DL-GPM (Fig. 4c and d) and single-layer graphene resonators (Fig. 4e and f). As shown in Fig. 4d and 4f, the resonant peaks of the modes A_2 and B_1 exhibit significantly smaller shifts of approximately $\Delta f = 0.09$ and 0.04 THz (blue-triangle and blue-star lines in Fig. 4g, respectively) when ε changes from 1 to 3, compared to the APC mode (red line in Fig. 4g).

To further quantitatively characterize the sensitivity of the APC mode to changes in the dielectric environment, we introduce a sensitivity (S) and figure of merit (FOM), defined as $S = \frac{\Delta f}{\Delta n}$, and $FOM = \frac{S}{FWHM}$, where Δf is the spectral shift, Δn is the change in refractive index, and FWHM is the full width at half-maximum of the resonant peak [45]. The calculated S and FOM of the APC mode ($S = 2.46$ THz/RIU, $FOM = 4.1$) is respectively 9 times and 10.5 times higher than that of the M_1 mode in the 2D t-DL-GPM, 20 times and 22 times higher than that of the A_2 mode in the untwisted DL-GPM, 45 times and 48 times higher than that of the B_1 mode in the single-layer graphene resonators. We further compared S and FOM of the APC mode in 2D t-DL-GPM with those of established THz sensing configurations (see Supplementary Table 1 for details) [46–49]. The APC mode outperforms the conventional split-ring resonator ($S \sim 0.11$ THz/RIU, $FOM \sim 0.14$) THz sensing configurations [48, 50], and achieves comparable performance to specifically designed quasi-bound state in the continuum (QBIC) based metasurfaces ($S \sim 0.42$ THz/RIU, $FOM \sim 6.5$) [49], despite operating with analyte volumes three orders of magnitude smaller ($100 \times 500 \times 5 \text{ nm}^3 \sim 280$ Ferritin protein molecules [51] and ~ 3500 bovine serum albumin molecules [52]). While the FOM remains lower than that from visible-range propagating surface plasmons in gold arrays or Mie resonances in dielectric metasurfaces due to inherent localized surface plasmon resonance (LSPR) losses [53, 54], like other LSPR based systems [55, 56], the APC mode's combination of exceptional sensitivity (2.46 THz/RIU) and strong field confinement in the nanocavity enables breakthrough detection capabilities at deep nanometer scales, positioning t-DL-GPM as a transformative platform for next-generation THz sensing applications.

Plasmonic properties in the transition from 2 to 1D t-DL-GPMs

We note that the absorption efficiency of the 2D t-DL-GPMs contributed by a single unit cell is too small to be detected using a conventional THz far-field spectrometer (such as, Fourier-transform infrared spectroscopy, FTIR). In principle, scattering-type scanning near-field optical microscopy (s-SNOM) could be employed to characterize the sensing performance, while the plasmon modes will be more complicated under s-SNOM tip excitation compared to far-field excitation, due to the tip's ability to generate higher momentum components [57, 58]. Herein we propose enhancing the absorption efficiency by leveraging the collective near-field coupling effects between unit cells in a periodic array. To explore this further, we investigated the influence of the inter-unit-cell distance d on the plasmonic properties of the 2D t-DL-GPMs (schematic shown in the upper and middle panels of Fig. 5a). As illustrated in Fig. 5b, the absorption efficiency increases significantly by a factor of 47 for the M_1 mode and 32 for the APC mode (M_4) (see Supplementary Figure S3) when d is reduced from

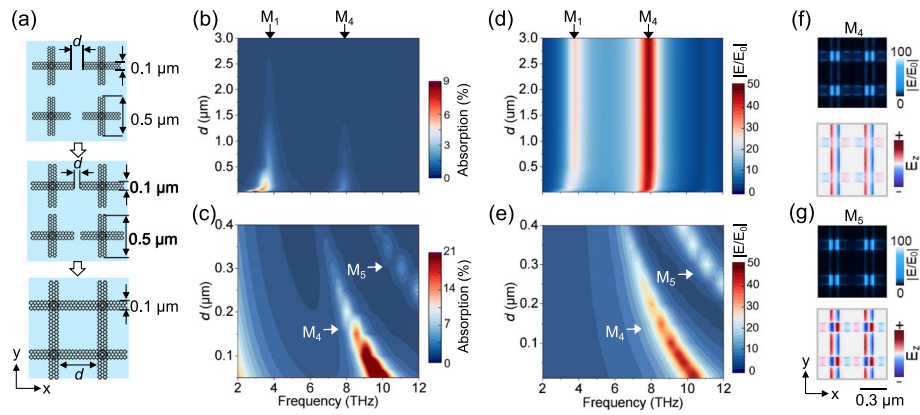


Fig. 5 Plasmonic properties in the transition from 2D to 1D t-DL-GPMs. **a** Schematics of the 2D t-DL-GPMs (upper and middle panels) and 1D t-DL-GPM (lower panel). The upper and middle panels depict the rectangle resonators with large and small inter-unit distance d , respectively. **b** and **c** Absorption spectra of the 2D t-DL-GPM and 1D t-DL-GPM as a function of d , respectively. **d** and **e** Average electric-field spectra of the 2D t-DL-GPM and 1D t-DL-GPM as a function of d , respectively. **f** and **g** Electric field (upper panel: $|E_x/E_0|$ and lower panel: E_z) distributions on the xy -plane for the M_4 (f) and M_5 (g) modes in 1D t-DL-GPM. The structural parameters: $W = 0.1 \mu\text{m}$, $L = 0.5 \mu\text{m}$, $h = 5 \text{ nm}$, $\theta = 90^\circ$.

$3.0 \mu\text{m}$ to $0.01 \mu\text{m}$. Notably, the resonant peaks of the M_1 and APC modes remain nearly constant as d decreases from $3.0 \mu\text{m}$ to $0.5 \mu\text{m}$ but exhibit red shifts with further reduction of d to $0.01 \mu\text{m}$. These observations are attributed to the enhanced near-field coupling between unit cells, analogous to the plasmon modes observed in metallic nanoparticle arrays and aggregates [59, 60]. The APC mode (M_4) demonstrates a larger electric field enhancement compared to the M_1 mode and shows minimal dependence on d . Additionally, the electric field enhancement for both modes remains stable as d changes from $3.0 \mu\text{m}$ to $0.5 \mu\text{m}$ but gradually decreases and then increases when d is further reduced to $0.01 \mu\text{m}$ (Fig. 5d). This phenomenon can be explained by the shift of the enhanced field region from the spacer between vertical resonators to the gap between horizontal resonators (see Supplementary Figure S3), resulting from the increased near-field coupling between horizontal resonators as d decreases.

Although the absorption of the APC mode increases, it remains insufficient for far-field FTIR spectroscopy experiments. To address this limitation, we propose 1D t-DL-GPMs, as illustrated in the lower panel of Fig. 5a. Figure 5c and e present the absorption spectra and electric field contour maps as functions of the inter-ribbon distance d , with the ribbon width W fixed at $0.1 \mu\text{m}$. Two distinct plasmon modes are observed in this configuration. The mode around 9 THz corresponds to the APC mode (M_4), as previously discussed. However, its electric field distribution (Fig. 5f) exhibits a dipolar characteristic on the upper ribbon and propagating plasmons on the lower ribbon, differing from the 2D t-DL-GPM due to the infinite extent of the lower ribbon along the x -direction. In contrast, the mode around 11 THz arises from propagating plasmons on both the top and lower ribbons (Fig. 5g). Notably, the absorption efficiency of the M_4 mode significantly increases as d decreases, reaching 21% at $d = 0.01 \mu\text{m}$, which is 12 times higher than that of the 2D t-DL-GPM under the same d . Additionally, the APC mode (M_4) maintains strong electric field enhancement

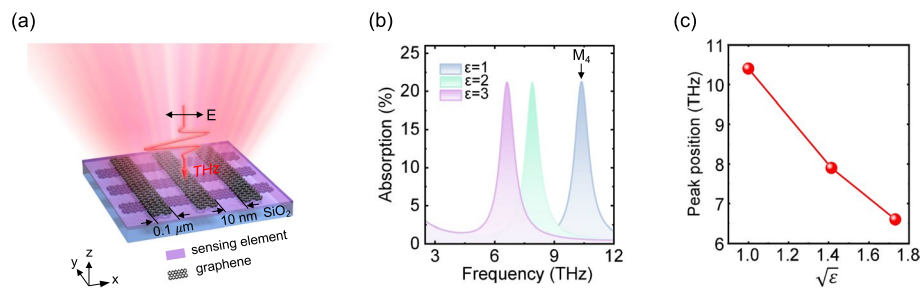


Fig. 6 Dependence of the absorption efficiency of 1D t-DL-GPM upon ϵ . **a** Schematic of 1D t-DL-GPM with $W = 0.1 \mu\text{m}$, $\theta = 90^\circ$, $d = 10 \text{ nm}$, $h = 5 \text{ nm}$, separated by sensing elements, represented by purple rectangle. **b** Absorption for the configuration in (a): $\epsilon = 1$, light-blue curve; $\epsilon = 2$, green curve; $\epsilon = 3$, pink curve. **c** Peak position depending on ϵ of the sensing element

(Fig. 5e), demonstrating its potential for high-performance sensing applications. These results highlight the advantages of the 1D t-DL-GPMs configuration in enhancing absorption efficiency, offering a promising platform for advanced THz sensing by far-field spectroscopy.

THz refractive index sensor with the 1D t-DL-GPMs

To demonstrate the sensing capability of 1D t-DL-GPMs, we investigated the spectral shift of the APC mode (M_4) as a function of ϵ in the spacer, as illustrated in Fig. 6a. Figure 6b shows the relationship between the spectral shift of the APC mode (M_4) on ϵ in a 5 nm thick spacer. The resonant peak exhibits a significant shift of $\Delta f = 3.8 \text{ THz}$ as ϵ changes from 1 to 3, demonstrating a linear relationship between the resonant peak position and ϵ (Fig. 6c). The corresponding FOM reaches 6.4, which is 1.5 times the FOM achieved by the 2D t-DL-GPM in Fig. 4b. These findings highlight the superior sensing capabilities of 1D t-DL-GPMs over its 2D counterpart in sensing applications.

Enhanced THz molecular vibrational fingerprints with APC mode in the 1D t-DL-GPMs

In addition to refractive index sensing, the strong plasmonic field confinement within the spacer, enabled by the APC mode, can be utilized to enhance THz vibrational spectroscopy for nanoscale molecules. This enhancement can be measured using FTIR or THz time-domain spectroscopy (THz TDS). To demonstrate the capability of fingerprint spectrum sensing, γ -aminobutyric acid (GABA, $\text{C}_7\text{H}_{15}\text{NO}_3$, Fig. 7a), a key biomarker molecule for characterizing glioma disease symptoms was selected as the analyte [61]. GABA exhibits two distinct vibrational bands at 1.01 THz and 1.51 THz, which are clearly reflected in its dielectric function, as shown in Fig. 7e. To investigate the vibrational sensing potential of 1D t-DL-GPMs, we tuned the resonant peak of the plasmon mode to align with the frequency range of GABA's absorption vibrations. This is achieved hereby increasing the geometric parameters of the graphene ribbons.

To assess the sensitivity of the proposed sensing schemes, we compared three distinct configurations, as illustrated in Fig. 7: (1) a single-layer graphene ribbon metasurface with a 5 nm thick GABA film atop the graphene (Fig. 7b); (2) a 1D untwisted DL-GPM, where a 5 nm thick GABA film is inserted into the spacer between the two vertical graphene ribbons (Fig. 7c); and (3) 1D t-DL-GPM with a 90-degree twist angle, with a 5 nm GABA film placed in the spacer between the two vertical ribbons (Fig. 7d).

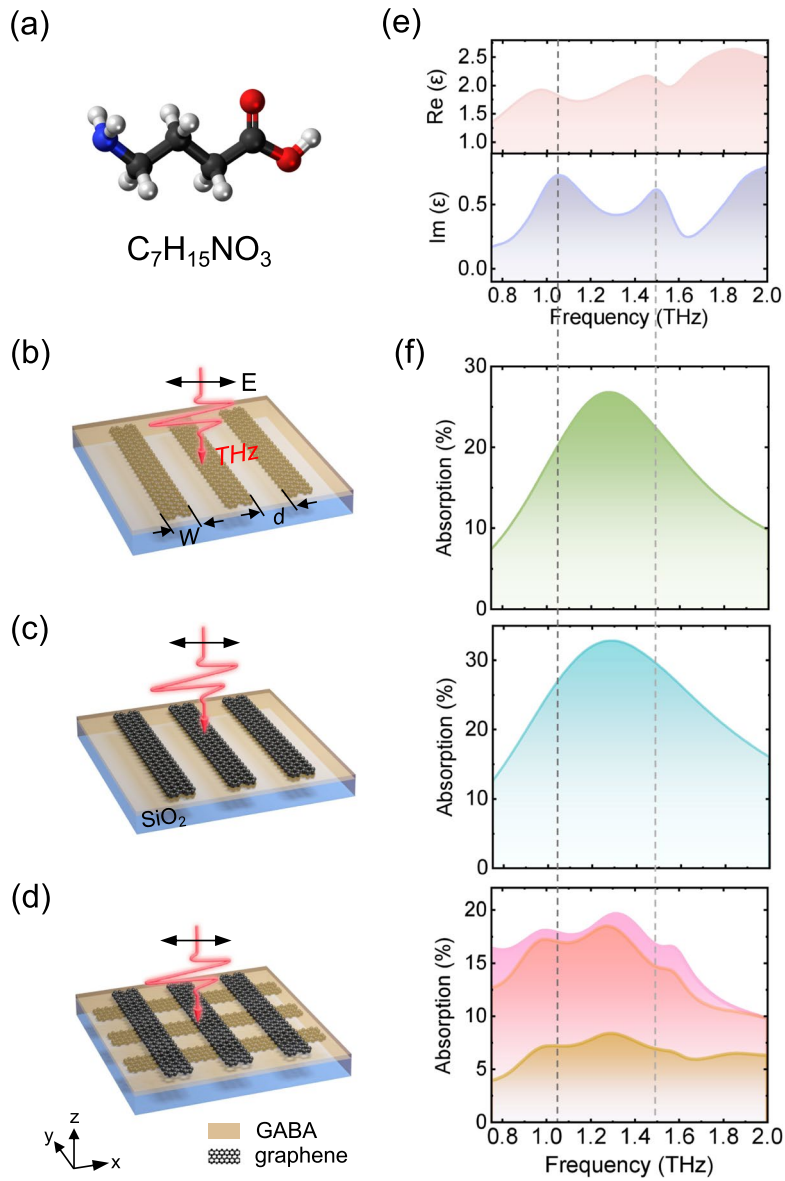


Fig. 7 1D t-DL-GPMs for THz sensing of molecular vibrational fingerprints. **a–d** Schematics depict: **(a)** molecular structures, **(b)** a single-layer graphene ribbon metasurface with a 5 nm molecular film on top of graphene sheet. The graphene ribbon has a width of $W = 2.39 \mu\text{m}$, distance of $d = 0.1 \mu\text{m}$ with a E_F of 0.455 eV. **c** An untwisted DL-GPM, featuring a 5 nm thick molecular film in the spacer. The parameters of the graphene ribbons are $W = 4.5 \mu\text{m}$, $d = 0.1 \mu\text{m}$ and $E_F = 0.455 \text{ eV}$. **d** A 90-degree 1D t-DL-GPM, also incorporating a few nm thick molecular film in the spacer. **e** The real part (upper panel) and imaginary part (lower panel) of the dielectric permittivity ϵ_{GABA} of the GABA molecule as a function of frequency. **f** Absorption spectra for the configurations shown in **(b)** (upper panel), **(c)** (middle panel), and **(d)** (lower panel). The pink, red and brown curves correspond to the configurations of **(d)** with $W = 0.65 \mu\text{m}$, $h = 5 \text{ nm}$, $d = 100 \text{ nm}$ and $E_{F,\text{upper}} = E_{F,\text{lower}} = 0.455 \text{ eV}$, $W = 0.3 \mu\text{m}$, $h = 1 \text{ nm}$, $d = 10 \text{ nm}$, $E_{F,\text{upper}} = 0.455 \text{ eV}$ and $E_{F,\text{lower}} = 0.25 \text{ eV}$, and $W = 0.2 \mu\text{m}$, $h = 1 \text{ nm}$, $d = 10 \text{ nm}$, $E_{F,\text{upper}} = 0.455 \text{ eV}$ and $E_{F,\text{lower}} = 0.05 \text{ eV}$, respectively. $E_{F,\text{upper}}$ and $E_{F,\text{lower}}$ is the Fermi level of upper and lower graphene ribbons

Their corresponding absorption spectra are presented in Fig. 7f. The spectrum for the single-layer graphene ribbon array fails to reveal the vibrational band signatures of GABA, highlighting the challenges faced by conventional plasmon modes in detecting

small-volume molecules. Similarly, the plasmon modes in the 1D untwisted DL-GPM (Fig. 7c) do not significantly enhance the visibility of the target molecules, as evidenced by the dark-green line in Fig. 7f, which shows no appreciable features. It is worthwhile to point out the absorption peaks in these two cases corresponding to the optical dipolar mode, which is induced by the strong near-field coupling between two horizontal ribbons when the distance d is small (e.g. 100 nm) [62]. The far-field excitation efficiency of the acoustic plasmon mode in the case of the untwisted DL-GPM is extremely low due to the near-zero scattering dipole moment, analogous a dark mode [62]. For the 1D t-DL-GPM, however, a dramatic improvement is observed, supporting the APC mode (Fig. 7d). In this case, the spectra are strongly influenced by the vibrational bands, with their fingerprints clearly identifiable in the spectra of the APC mode (pink curve in the lower panel of Fig. 7f). Due to Fano interference between the APC mode and the molecular vibrational bands, the vibrational features do not appear as distinct peaks in the spectra, unlike in the imaginary part of the dielectric function. This phenomenon is consistent with previous observations, such as plasmon modes in gold antennas enhancing IR fingerprints of molecules [63]. The remarkable vibrational signatures detected for an extremely thin molecular film underscore the potential of t-DL-GPM as a highly promising platform for enhanced THz absorption spectroscopy applications.

It is crucial to emphasize the exceptionally small mode volume of the APC mode in the 1D t-DL-GPM, which is the key contribution to the sensing performance. This normalized mode volume $\left(V'_m\right)$ of an APC mode (the pink curve in Fig. 7) reaches $1 \times 10^{-11} \lambda_0^3$. Meanwhile, around 70% of the energy remains confined in the interlayer 3D cavity (Supplementary Figure S4a-c). Further reduction is achievable by tuning the graphene's Fermi level (E_F), horizontal distance (d) of two ribbons and spacer thickness (h). Graphene's atomic thickness enables easy carrier density (and thus E_F) control via gating (Fig. 1a), granting tunability advantages. As demonstrated in Fig. 7f (red curve), lowering the bottom graphene's E_F to 0.25 eV (while optimizing geometry parameters) enhances molecular vibrational fingerprints. By further reducing $E_{F, \text{lower}}$ to 0.05 eV, W to 0.2 μm , d to 10 nm and h to 1 nm, the mode volume $\left(V'_m\right)$ (brown curve) shrinks tens of times (reaching $1 \times 10^{-13} \lambda_0^3$, achieving single monolayer GABA detection (Supplementary Figure S4d-e). This achievable thickness resolution is intrinsically linked to molecular properties to some extent, most notably the vibrational mode strength. Such ultra-small volumes could hold great promise for achieving strong coupling and even pave the way for single-molecule detection at THz frequency [64, 65]. Our theoretical calculations confirm that strong coupling between 1D t-DL-GPM and THz molecules can indeed be achieved through graphene loss reduction (i.e., mobility enhancement) [12, 66], as detailed in Supplementary Figure S5. While these THz sensing and strong coupling platforms hold tremendous scientific potential, their experimental implementation via THz far-field spectroscopy faces challenges in synthesizing and fabricating high-quality, large-area t-DL-GPM. Critical issues such as high breakage rates during mechanical transfer, mobility degradation from interface contamination, and increased defect densities due to PMMA residue could substantially degrade sample quality. Additionally, THz far-field spectroscopic characterization shows suboptimal performance by its relatively lower signal-to-noise ratio as compared to optical frequencies and the immature state of optical microscopy—especially below 3 THz, which consequently imposes more

stringent requirements on sample quality and dimensions. These technical limitations underscore the importance of this work as a theoretical foundation for future experimental investigations. Alternatively, THz s-SNOM—an advanced characterization technique enabling nanoscale spatial resolution [21, 67, 68]—can overcome sample size constraints. When combined with recent advances in alternative high-quality THz polaritonic materials (e.g., Ag_2Te) [22], this THz nanoscopy based methodology holds exceptional potential to propel the field forward. Additionally, we note that when the interlayer spacing approaches 10 nm, quantum effects (e.g., nonlocal response, tunneling) may begin to emerge [69, 70]. While these effects may introduce minor modifications to the resonance peak positions and intensities of plasmon modes, they are not expected to substantially alter the conclusions of this work. Future work will explore these quantum phenomena systematically.

Conclusion

We systematically investigated the plasmonic properties of twisted double-layer graphene plasmonic metasurfaces (t-DL-GPMs) in both 1D and 2D configurations. These structures were revealed to support a series of acoustic plasmon modes in the THz frequency range, arising from the coupling between upper and lower graphene resonators. Particularly, the acoustic plasmon cavity (APC) mode — formed through the vertical coupling of different-order plasmon modes on upper and lower resonators—exhibits remarkable electric-field enhancement and ultrastrong field confinement, achieving an exceptionally small mode volume down to $10^{-13} \lambda_0^3$. This outstanding performance originates from the 3D nanocavity enabled by the twisting manipulation of the graphene resonators. Our study reveals that the field enhancement and absorption efficiency of the APC mode are highly dependent on both the twist angle and near-field coupling between lateral resonators. We also demonstrate that t-DL-GPMs can function as highly sensitive sensors for ultrasmall analyte volumes. The sensitivity of the APC mode significantly surpasses that of conventional optical modes in single-layer graphene metasurfaces and acoustic plasmon modes in untwist DL-GPMs, with FOM that are 48 and 22 times higher, respectively. Notably, our results reveal that the 1D t-DL-GPM enhances THz molecular vibrational fingerprints with unprecedented 1 nm thickness sensitivity. These findings not only hold significant importance for THz sensing applications of nanoscale molecules and the study of ultrastrong light-matter interactions^[59,68], but also establish a robust platform for the developing advanced THz optoelectronic devices.

Methods

Full Wave Simulations

In this study, we performed numerical simulations using the commercial finite-element software COMSOL Multiphysics (version 6.0). The computational domain was configured with Floquet periodic boundary conditions along both x- and y-axes to simulate an infinite periodic array in free space, while a perfectly matched layer (PML) was implemented along the z-direction to eliminate unwanted reflections [71]. The metasurface was illuminated by a linearly x-polarized THz plane wave propagating along the z-axis. Graphene's electro-optical properties were modeled through surface boundary conditions, incorporating its frequency-dependent surface conductivity($\sigma_g(\omega)$) calculated

within the local random-phase approximation framework [72]. The silicon oxide substrate was characterized by a constant relative permittivity of $\epsilon=4$ [73]. In our theoretical framework, the twisted double-layer graphene system is modeled as two rotationally isotropic conductive sheets with a relative twist angle θ , while the untwisted configuration ($\theta=0$) represents the aligned case.

Mode volume (V_m) calculations

In this work, the mode volume is calculated by using $V_m = \frac{\int \epsilon(r) |E(r)|^2 d^3r}{\max[\epsilon(r) |E(r)|^2]}$, where $E(r)$

and $\epsilon(r)$ is the electric field and dielectric permittivity at the spatial location r . The integral volume operates to the cavity region (overlap area of double-layer graphene). This is a classical definition that widely used in the literature [74].

Dispersive corrected mode volume (V'_m) calculation

Considering the dispersive nature of graphene, the mode volume is further corrected and calculated via [75]:

$$V'_m = \frac{\int \frac{\partial(\omega\epsilon(r,\omega))}{\partial\omega} |E(r)|^2 d^3r}{\max\left[\frac{\partial(\omega\epsilon(r,\omega))}{\partial\omega} |E(r)|^2\right]} \quad (1)$$

where ω is the angle frequency, r is the spatial location in air/graphene, $\epsilon(r, \omega)$ is the frequency-dependent dielectric permittivity at the spatial location r . Considering the distinct cavity regimes for mode volume calculations arising from (i) the interlayer spacer and (ii) the top and bottom graphene layers themselves, with position-independent permittivity, V'_m is derived as the following refined expression:

$$V'_m = \frac{\int (\epsilon_a(\omega) |E(r)|^2 d^3r_a + \frac{\partial(\omega\epsilon_g(\omega))}{\partial\omega} |E(r)|^2 h d^2r_g)}{\max\left[\left(\epsilon_a(\omega) + \frac{\partial(\omega\epsilon_g(\omega))}{\partial\omega}\right) |E(r)|^2\right]} \quad (2)$$

where $h=0.33$ nm is thickness of monolayer graphene, $\epsilon_a(\omega)$ and $\epsilon_g(\omega)$ are dielectric permittivity of the interlayer spacer and graphene, respectively. Here, graphene is modeled as a 2D conductive surface with conductivity $\sigma_g(\omega)$, which satisfies $\sigma_g(\omega) = -i\omega h \epsilon_g(\omega)/4\pi$ [72]. Consequently, the volume integral over graphene reduces to a surface integral multiplied by its effective thickness h . Note that for the non-dispersive materials, $V'_m=V_m$.

Energy portion (η) calculations

The portion of electric-field intensity is introduced to evaluate the capability of field concentration and sensing of the acoustic plasmon cavity mode. It is calculated by using

$$\eta = \frac{\int_{\text{cavity}} \epsilon_a(\omega) |E(r)|^2 d^3r_a}{\int_{\text{all}} \frac{\partial(\omega\epsilon(r,\omega))}{\partial\omega} |E(r)|^2 d^3r} \quad (3)$$

Here, the numerator integrals correspond to the cavity region (overlapped dielectric spacer between graphene layers) and denominator integrals correspond to the complete graphene structure in Fig. 2 and one unit cell in Fig. 7d, respectively. For the energy

portion and mode volume calculations in Fig. 2, we set $\varepsilon_a(\omega) = 1$ (air). In Fig. 7d, the dielectric permittivity of GABA molecules is fixed at their 1.03 THz value (left vibrational absorption peak), justified by: (1) their negligible dispersion characteristics, and (2) our exclusive focus on this specific vibrational absorption frequency.

Supplementary Information

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Supplementary Material 1

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Authors' contributions

S.C. conceived the idea, supervised the project, wrote the manuscript and acquired the funds. H.Z. performed the simulations and data analysis, assisted with manuscript composition. Y.Z. and S.Z. acquired the funds and co-supervised the project. P.L., X.Y., W.W. and G.F. discussed the results and assisted with manuscript composition.

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Data availability

The data and the relevant methods are available on request from the corresponding authors.

Declarations

Ethics approval and consent to participate

Not applicable

Competing interests

The authors declare no competing interests.

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References

1. Ferguson B, Zhang X-C. Materials for terahertz science and technology. *Nat Mater*. 2002;1:26–33.
2. Niessen KA, Xu M, George DK, et al. Protein and RNA dynamical fingerprinting. *Nat Commun*. 2019;10(1):1026.
3. Tonouchi M. Cutting-edge terahertz technology. *Nat Photon*. 2007;1:97–105.
4. Jin X, Aglieri V, Jeong YG, et al. Enhanced terahertz spectroscopy of a monolayer transition metal dichalcogenide. *Adv Funct Mater*. 2025;24:2419841.
5. Zhang Z, Wang Z, Zhang C, et al. Advanced terahertz refractive sensing and fingerprint recognition through metasurface-excited surface waves. *Adv Mater*. 2024;36:2308453.
6. Chen S, Autore M, Li J, et al. Acoustic graphene plasmon nanoresonators for field-enhanced infrared molecular spectroscopy. *ACS Photonics*. 2017;4:3089–97.
7. Rodrigo D, Limaj O, Janner D, et al. Mid-infrared plasmonic biosensing with graphene. *Science*. 2015;349:165–8.
8. Toma A, Tuccio S, Prato M, et al. Squeezing terahertz light into nanovolumes: Nanoantenna enhanced terahertz spectroscopy (NETS) of semiconductor quantum dots. *Nano Lett*. 2014;15:386–91.
9. Baumberg JJ, Aizpurua J, Mikkelsen MH, et al. Extreme nanophotonics from ultrathin metallic gaps. *Nat Mater*. 2019;18:668–78.
10. Xu H, Aizpurua J, Käll M, et al. Electromagnetic contributions to single-molecule sensitivity in surface-enhanced Raman scattering. *Phys Rev E*. 2000;62:4318–24.
11. Zhang R, Zhang Y, Dong ZC, et al. Chemical mapping of a single molecule by plasmon-enhanced Raman scattering. *Nature*. 2013;498:82–6.
12. Chikkaraddy R, de Nijs B, Benz F, et al. Single-molecule strong coupling at room temperature in plasmonic nanocavities. *Nature*. 2016;535:127–30.
13. Chen W, Zhang S, Deng Q, et al. Probing of sub-picometer vertical differential resolutions using cavity plasmons. *Nat Commun*. 2018;9:801.
14. Li C-Y, Duan S, Wen B-Y, et al. Observation of inhomogeneous plasmonic field distribution in a nanocavity. *Nat Nanotechnol*. 2020;15:922–6.

15. Li J, Wu D, Li J, et al. Ultrasensitive plasmon-enhanced infrared spectroelectrochemistry. *Angew Chem Int Edit*. 2024;63:e202319246.
16. Zhang X, Xu Q, Xia L, et al. Terahertz surface plasmonic waves: a review. *Adv Photon*. 2020;2:014001.
17. Aupiais I, Grasset R, Guo T, et al. Ultrasensitive and tunable terahertz surface plasmon cavities at the ultimate plasmonic limit. *Nat Commun*. 2023;14:7645.
18. Keller J, Scalari G, Cibella S, et al. Few-electron ultrastrong light-matter coupling at 300 GHz with nanogap hybrid LC microcavities. *Nano Lett*. 2017;17:7410–5.
19. Chen J, Badioli M, Alonso-González P, et al. Optical nano-imaging of gate-tunable graphene plasmons. *Nature*. 2012;487:77–81.
20. Fei Z, Rodin AS, Andreev GO, et al. Gate-tuning of graphene plasmons revealed by infrared nano-imaging. *Nature*. 2012;487:82–5.
21. Chen S, Bylinkin A, Wang Z, et al. Real-space nanoimaging of THz polaritons in the topological insulator Bi_2Se_3 . *Nat Commun*. 2022;13(1):1374.
22. Chen S, Leng PL, Konečná A, et al. Real-space observation of ultraconfined in-plane anisotropic acoustic terahertz plasmon polaritons. *Nat Mater*. 2023;22:860–6.
23. Alonso-González P, Nikitin AY, Gao Y, et al. Acoustic terahertz graphene plasmons revealed by photocurrent nanoscopy. *Nat Nanotechnol*. 2017;12:31–5.
24. Menabde SG, Heiden JT, Cox JD, et al. Image polaritons in van der Waals crystals. *Nanophotonics*. 2022;11:2433–52.
25. Stauber T, Gómez-Santos G. Plasmons in layered structures including graphene. *New J Phys*. 2012;14:105018.
26. Principi A, Asgari R, Polini M. Acoustic plasmons and composite hole-acoustic plasmon satellite bands in graphene on a metal gate. *Solid State Commun*. 2011;151:1627–30.
27. Gu X, Lin IT, Liu J-M. Extremely confined terahertz surface plasmon-polaritons in graphene-metal structures. *Appl Phys Lett*. 2013;103:071103.
28. Lee I-H, Yoo D, Avouris P, et al. Graphene acoustic plasmon resonator for ultrasensitive infrared spectroscopy. *Nat Nanotechnol*. 2019;14:313–9.
29. Menabde SG, Lee IH, Lee S, et al. Real-space imaging of acoustic plasmons in large-area graphene grown by chemical vapor deposition. *Nat Commun*. 2021;12:938.
30. Oh S-H, Altug H, Jin X, et al. Nanophotonic biosensors harnessing van der Waals materials. *Nat Commun*. 2021;12:3824.
31. Sunku SS, Ni GX, Jiang BY, et al. Photonic crystals for nano-light in moiré graphene superlattices. *Science*. 2018;362:1153–6.
32. Huang T, Tu X, Shen C, et al. Observation of chiral and slow plasmons in twisted bilayer graphene. *Nature*. 2022;605:63–8.
33. Hu F, Das SR, Luan Y, et al. Real-space imaging of the tailored plasmons in twisted bilayer graphene. *Phys Rev Lett*. 2017;119:247402.
34. Hesp NCH, Torre I, Rodan-Legrain D, et al. Observation of interband collective excitations in twisted bilayer graphene. *Nat Phys*. 2021;17:1162–8.
35. Cavicchi L, Torre I, Jarillo-Herrero P, et al. Theory of intrinsic acoustic plasmons in twisted bilayer graphene. *Phys Rev B*. 2024;110(4):045431.
36. Zhang H, Fan X, Wang D, et al. Electric field-controlled damping switches of coupled dirac plasmons. *Phys Rev Lett*. 2022;129:237402.
37. Zhou C-L, Wu X-H, Zhang Y, et al. Polariton topological transition effects on radiative heat transfer. *Phys Rev B*. 2021;103:155404.
38. Liu Z, Zhang Z, Zhou F, et al. Dynamically tunable electro-optic switch and multimode filter based on twisted bilayer graphene strips. *J Opt*. 2021;23:025104.
39. Cui W, Wang Y, Xue J, et al. Terahertz sensing based on tunable fano resonance in graphene metamaterial. *Results Phys*. 2021;31:104994.
40. Moreno Á, Cavicchi L, Wang X, et al. Twisted bilayer graphene for enantiomeric sensing of chiral molecules. 2024, <https://arxiv.org/abs/2409.05178>.
41. Christensen J, Manjavacas A, Thongrattanasiri S, et al. Graphene plasmon waveguiding and hybridization in individual and paired nanoribbons. *ACS Nano*. 2012;6:431–40.
42. Nikitin AY, Alonso-González P, Vélaz S, et al. Real-space mapping of tailored sheet and edge plasmons in graphene nanoresonators. *Nat Photon*. 2016;10:239–43.
43. Wu C, Duan Y, Yu L, et al. In-situ observation of silk nanofibril assembly via graphene plasmonic infrared sensor. *Nat Commun*. 2024;15:4643.
44. Chen S, Zhang Y, Shih T-M, et al. Plasmon-induced magnetic resonance enhanced raman spectroscopy. *Nano Lett*. 2018;18:2209–16.
45. Lyu J, Huang L, Chen L, et al. Review on the terahertz metasensor: from featureless refractive index sensing to molecular identification. *Photonics Res*. 2024;12:194–217.
46. Liu W, Zhou X, Zou S, et al. High-sensitivity polarization-independent terahertz Taichi-like micro-ring sensors based on toroidal dipole resonance for concentration detection of A β protein. *Nanophotonics*. 2023;12:1177–87.
47. Zhang J, Mu N, Liu L, et al. Highly sensitive detection of malignant glioma cells using metamaterial-inspired THz biosensor based on electromagnetically induced transparency. *Biosens Bioelectron*. 2021;185:113241.
48. Tao H, Strikwerda AC, Liu M, et al. Performance enhancement of terahertz metamaterials on ultrathin substrates for sensing applications. *Appl Phys Lett*. 2010;97:261909.
49. Liu B, Peng Y, Jin Z, et al. Terahertz ultrasensitive biosensor based on wide-area and intense light-matter interaction supported by QBC. *Chem Eng J*. 2023;462:142347.
50. Lan F, Luo F, Mazumder P, et al. Dual-band refractometric terahertz biosensing with intense wave-matter-overlap microfluidic channel. *Biomed Opt Express*. 2019;10:3789–99.
51. Amenabar I, Poly S, Nuansing W, et al. Structural analysis and mapping of individual protein complexes by infrared nanospectroscopy. *Nat Commun*. 2013;4:2890.

52. Röcker C, Pötzl M, Zhang F, et al. A quantitative fluorescence study of protein monolayer formation on colloidal nanoparticles. *Nat Nanotechnol.* 2009;4:577–80.
53. Liu B, Chen S, Zhang J, et al. A Plasmonic sensor array with ultrahigh figures of merit and resonance linewidths down to 3 nm. *Adv Mater.* 2018;30:e1706031.
54. Qiu S, Zhang H, Shi Z, et al. Ultrasensitive refractive index sensing based on hybrid high-Q metasurfaces. *J Phys Chem C.* 2023;127:8263–70.
55. Zhang S, Bao K, Halas NJ, et al. Substrate-induced fano resonances of a plasmonic nanocube: a route to increased-sensitivity localized surface plasmon resonance sensors revealed. *Nano Lett.* 2011;11:1657–63.
56. Verellen N, Van Dorpe P, Huang C, et al. Plasmon line shaping using nanocrosses for high sensitivity localized surface plasmon resonance sensing. *Nano Lett.* 2011;11:391–7.
57. Dolado I, Maciel-Escudero C, Nikulina E, et al. Remote near-field spectroscopy of vibrational strong coupling between organic molecules and phononic nanoresonators. *Nat Commun.* 2022;13:6850.
58. Bylinkin A, Schnell M, Autore M, et al. Real-space observation of vibrational strong coupling between propagating phonon polaritons and organic molecules. *Nat Photonics.* 2020;15:197–202.
59. Le F, Brandl DW, Urzhumov YA, et al. Metallic nanoparticle arrays: a common substrate for both surface-enhanced Raman scattering and surface-enhanced infrared absorption. *ACS Nano.* 2008;2:707–18.
60. Chen W, Zhang S, Kang M, et al. Probing the limits of plasmonic enhancement using a two-dimensional atomic crystal probe. *Light Sci Appl.* 2018;7:56.
61. Liu B, Peng Y, Hao Y, et al. Ultra-wideband terahertz fingerprint enhancement sensing and inversion model supported by single-pixel reconfigurable graphene metasurface. *Photonix.* 2024;5:10.
62. Halas NJ, Lal S, Chang W-S, et al. Plasmons in strongly coupled metallic nanostructures. *Chem Rev.* 2011;111:3913–61.
63. Neubrech F, Huck C, Weber K, et al. Surface-enhanced infrared spectroscopy using resonant nanoantennas. *Chem Rev.* 2017;117:5110–45.
64. Wang YH, Zheng S, Yang WM, et al. In situ Raman spectroscopy reveals the structure and dissociation of interfacial water. *Nature.* 2021;600:81–5.
65. Wen BY, Wang JY, Shen TL, et al. Manipulating the light-matter interactions in plasmonic nanocavities at 1 nm spatial resolution. *Light Sci Appl.* 2022;11:235.
66. Damari R, Weinberg O, Krotkov D, et al. Strong coupling of collective intermolecular vibrations in organic materials at terahertz frequencies. *Nat Commun.* 2019;10:3248.
67. Zhang T, Chen S, Petkov PS, et al. Two-dimensional polyaniline crystal with metallic out-of-plane conductivity. *Nature.* 2025;638:411–7.
68. Zizlsperger M, Nerreter S, Yuan Q, et al. In situ nanoscopy of single-grain nanomorphology and ultrafast carrier dynamics in metal halide perovskites. *Nat Photon.* 2024;18:975–81.
69. Lundeborg MB, Gao Y, Asgari R, et al. Tuning quantum nonlocal effects in graphene plasmonics. *Science.* 2017;357:187–91.
70. Zhu W, Esteban R, Borisov AG, et al. Quantum mechanical effects in plasmonic structures with subnanometre gaps. *Nat Commun.* 2016;7:11495.
71. Li P, Wang H, Turup Z, et al. Efficient manipulation of plasmonic modes in single symmetry-breaking Ag nanocube. *Appl Surf Sci.* 2023;611:155650.
72. Stefan AM. (Ed.). *World Scientific handbook of metamaterials and plasmonics* (Vol. 4). Singapore: World Scientific Publishing Co. Pte. Ltd.; 2017.
73. Giustino F, Umari P, Pasquarello A. Dielectric effect of a thin SiO₂ interlayer at the interface between silicon and high-k oxides. *Microelectron Eng.* 2004;72:299–303.
74. Huang S, Ming T, Lin Y, et al. Ultrasmall mode volumes in plasmonic cavities of nanoparticle-on-mirror structures. *Small.* 2016;12:5190–9.
75. Li X, Smalley JST, Li ZT, Gu Q. Effective modal volume in nanoscale photonic and plasmonic near-infrared resonant cavities. *Appl Sci.* 2018;8:1646.

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