

Collective Modes in Multilayer Graphene/ α -RuCl₃ Heterostructures

Samuel L. Moore^{1,*}, Miguel Sánchez Sánchez², M. C. Strassbourg³, Y. Shao⁴, J. Pack¹, Y. Wang⁵,
D. J. Rizzo¹, B. S. Jessen⁶, Matthew Cothrine⁷, David G. Mandrus^{7,8}, Takashi Taniguchi⁹, Kenji Watanabe¹⁰,
K. S. Burch¹¹, C. R. Dean¹, J. Hone³, M. Fogler¹², A. J. Millis¹, A. Rubio¹³,
P. J. Schuck³, T. Stauber², and D. N. Basov¹

¹Physics Department, Columbia University, New York, New York, USA

²Instituto de Ciencia de Materiales de Madrid, ICMM-CSIC, Madrid, Spain

³Mechanical Engineering Department, Columbia University, New York, New York, USA

⁴Physics Department, Pennsylvania State University, University Park, Pennsylvania, USA

⁵Chemistry Department, Columbia University, New York, New York,

⁶Physics Department, Technical University of Denmark, Kongens Lyngby, Denmark

⁷Department of Materials Science and Engineering, University of Tennessee, Knoxville, Tennessee, USA

⁸Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA

⁹Research Center for Materials Nanoarchitectonics,

National Institute for Materials Science, 1-1 Namiki, Tsukuba, Japan

¹⁰Research Center for Electronic and Optical Materials,

National Institute for Materials Science, 1-1 Namiki, Tsukuba, Japan

¹¹Physics Department, Boston College, Boston, Massachusetts, USA

¹²Physics Department, University of California, San Diego, California, USA

¹³Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany



(Received 5 December 2024; revised 13 May 2025; accepted 23 July 2025;
published 21 October 2025; corrected 3 December 2025)

Collective modes in multilayer graphene, such as plasmons and phonons, exhibit sensitivity to displacement fields and interlayer coupling, distinguishing them from their counterparts in single-layer graphene. Here, we engineer collective modes in charge-transfer heterostructures composed of multilayer graphene and α -RuCl₃. In heterostructures with a single α -RuCl₃ interface, the charge transfer generates displacement fields up to 7 V/nm at the interface between α -RuCl₃ and the adjacent graphene layer—the highest value achieved through charge-transfer methods. As a result of the broken inversion symmetry, we discover enhanced nonlinear optical response and modified phonon selection rules. Conversely, we find that multilayer graphene sandwiched between two α -RuCl₃ flakes causes displacement fields to cancel. There, we achieve carrier densities as high as $8 \times 10^{13} \text{ cm}^{-2}$ in multilayer graphene and restore the phonon selection rules to their unperturbed state. Meanwhile, we demonstrate that plasmonic properties derive from the depletion of multiple valence bands. As a result of the quasilinear band dispersion, these “Dirac multiband plasmons” are relatively unaffected by displacement fields. On the other hand, the inverted heterostructure sequence—two multilayer graphene sheets encapsulating α -RuCl₃—activates significant alteration of the plasmons via interlayer plasmon-plasmon coupling. Hence, multilayer graphene and α -RuCl₃ heterostructures offer a gate-free platform for engineering collective modes derived from inversion symmetry and interlayer coupling.

DOI: [10.1103/tp3q-svws](https://doi.org/10.1103/tp3q-svws)

Subject Areas: Condensed Matter Physics, Optics,
Plasmonics

I. INTRODUCTION

Collective modes are ubiquitous throughout condensed matter physics, including phonons and plasmons. In multi

(N)-layer graphene (NLG), phonons and plasmon polaritons are robust collective modes with sensitivity to external controls inaccessible in single-layer graphene (SLG), e.g., displacement fields D [1–6] and interlayer coupling g [7–11]. Unfortunately, plasmonic properties of NLG under large carrier densities and displacement fields have been thus far unattainable as a result of incompatible device geometries. Likewise, electrostatically gated devices examining plasmon-plasmon interlayer coupling operate only at low carrier densities ($<10^{13} \text{ cm}^{-2}$) [10], thus limiting the frequency range of plasmonic applications.

*Contact author: slm2236@columbia.edu

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

Here, we utilize van der Waals (vdW) charge-transfer interfaces to engineer collective modes in NLG, achieved through the strategic use of interlayer coupling, displacement fields, and ultrahigh carrier densities. While these heterostructures lack *in situ* carrier tunability, their simple fabrication makes them compatible with novel experimental probes of collective mode in highly doped NLG. We consider both bilayer and trilayer graphene (BLG and TLG, respectively), the latter of which has yet to be experimentally examined in vdW charge-transfer heterostructures. We induce ultrahigh carrier densities and displacement fields by forming multi-interface vertical heterostructures of NLG with α -RuCl₃, the vdW material with the largest attainable work function thus far [12]. In single-interface charge-transfer heterostructures (NLG/R), we report nearly 3 times stronger displacement fields than obtained for BLG on the notable charge-transfer compound CrCl₃ [13]. Meanwhile, the net displacement field is absent in double-interface heterostructures (R/NLG/R) due to the symmetric carrier density profile in NLG.

In Fig. 1, we provide a generic overview on how displacement fields, multiband filling, and interlayer plasmon coupling impact the plasmons and phonons of NLG. Figures 1(a) and 1(b) first consider the plasmonic collective modes. In doped NLG, hybrid light/charge-carrier modes known as surface plasmon polaritons (SPPs) emerge at infrared frequencies that confine optical fields below the diffraction limit [12,14]. The SPP is described in terms of the optical conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$, shown in Fig. 1(a) for BLG on α -RuCl₃ (Appendix H). The SPP wavelength satisfies $\lambda_p \propto \mathcal{D}_W/\omega^2$, where ω is the optical excitation frequency and $\mathcal{D}_W \equiv (1/\pi) \int_0^{\omega_p} d\omega \sigma_1(\omega)$ is the Drude weight. The blue shaded area of Fig. 1(a) defines the frequency range $0 < \omega < \omega_D$ where interband absorption can be neglected. Meanwhile, the gray shaded area of Fig. 1(a) denotes the frequency range over which plasmons were experimentally interrogated in this work. Within this frequency window, losses are low (i.e., $\sigma_2 \approx \mathcal{D}_W/\omega \gg \sigma_1$), and thus the plasmons propagate over long distances [lower panel of Fig. 1(a)].

In Fig. 1(b), we consider the novel behaviors of plasmons that result from high carrier density, displacement fields, and interlayer coupling. First, we focus on the plasmonic properties stemming from multiple NLG valence bands $\beta = \{\text{VB1, VB2, } \dots\}$. Here, we may express the Drude weight as

$$\mathcal{D}_W \equiv \sum_{\beta} \mathcal{D}_W^{(\beta)} \propto \sum_{\beta} S_{\beta} |v_{\beta}|, \quad (1)$$

where v_{β} is a Fermi-surface-averaged velocity matrix element of band β and S_{β} is the corresponding circumference of the Fermi surface (see Appendix H). At ultrahigh carrier densities, the primary outcome of the large displacement

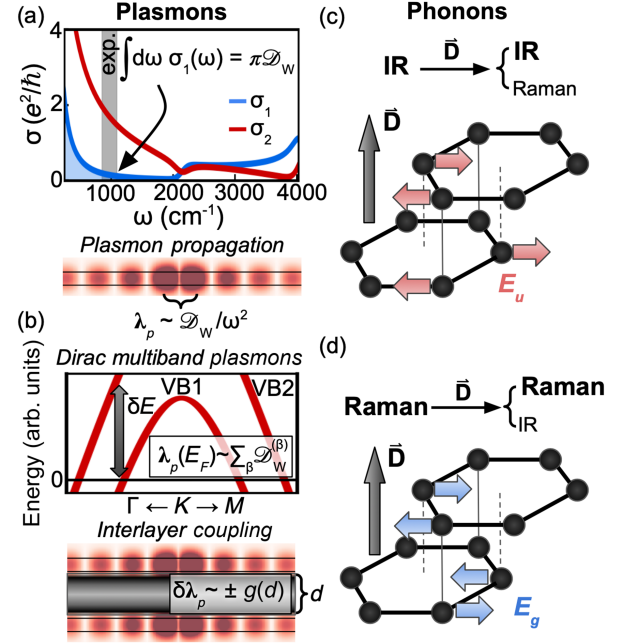


FIG. 1. Overview of collective modes in multilayer graphene at high carrier densities and displacement fields. Panel (a) plots the BLG/R optical conductivity $\sigma_1(\omega)$ and $\sigma_2(\omega)$. The blue shaded area defines the Drude weight, the frequency range where interband transitions are negligible. Meanwhile, the gray shaded area (“exp.”) denotes the frequency range where plasmons, with wavelength $\lambda_p \sim \mathcal{D}_W/\omega^2$, were studied. The upper panel of (b) sketches the BLG band structure for the Fermi energies E_F attained in this work. The Fermi level crosses both valence bands VB1 and VB2, resulting in a plasmon wavelength λ_p that depends on the Drude weights of both bands. Upon separating two NLG sheets by a spacer of thickness d , the lower panel in (b) suggests that the plasmon wavelength may be split by the amount $\delta\lambda_p \sim \pm g(d)$ in response to interlayer plasmon-plasmon coupling $g(d)$. Panels (c) and (d) illustrate two optical phonon modes in bilayer graphene. The infrared-active mode (E_u) corresponds to out-of-phase interlayer oscillations, as shown by red arrows in (c). Meanwhile, the Raman-active phonon (E_g) exhibits in-phase interlayer oscillations, indicated by the blue arrows in panel (d). Upon applying a displacement field D , the modes lose their distinct IR and Raman activity, denoted by $\text{IR} \rightarrow \{\text{IR}, \text{Raman}\}$.

field D is to modify both the band gap Δ and the splitting δE between the valence bands. As sketched in Fig. 1(b), these bands are approximately linear at the Fermi level E_F . For BLG, the corresponding Drude weights obey the Dirac-like form $\mathcal{D}_W^{(\beta)} \sim 4\sigma_0(|E_F| + \delta E_{\beta})/\pi$, where $\delta E_2 - \delta E_1 = \delta E$ and $\sigma_0 \equiv e^2/(4\hbar)$ is the universal graphene conductivity [8]. A similar expression is derived for TLG bands. As a result of the linear band dispersion, NLG plasmons become independent of displacement fields upon doping into all valence bands, resembling the plasmonic response of N decoupled

monolayers. We therefore term this unique collective mode a “Dirac multiband plasmon.” On the other hand, the bottom panel of Fig. 1(b) illustrates that plasmon-plasmon coupling provides an alternative tool for controlling plasmons, achieved by separating two plasmonic sheets by a distance $d \sim \lambda_p/(2\pi)$. Plasmon-plasmon coupling results in the mode splitting $\delta\lambda_p/\lambda_p \sim \pm g(d)$, where $g(d) \sim e^{-2\pi d/\lambda_p}$ is the coupling strength [10]. Therefore, the coupling between two NLG sheets is expected to produce confined plasmon propagation, even at high carrier densities.

Figures 1(c) and 1(d) illustrate that the strong displacement fields generated in NLG/R stacks are essential for controlling NLG optical phonons. Inversion symmetric BLG hosts one infrared-active optical phonon [E_u , Fig. 1(c)], corresponding to out-of-phase lattice oscillations between top and bottom layers [1,3]. However, the strong displacement field breaks inversion symmetry and alters the phonon selection rules, enabling weak infrared excitation of the symmetric mode (E_g) in Fig. 1(d). Conversely, inversion symmetry breaking also prescribes a small Raman activity to the antisymmetric mode E_u in Fig. 1(c). The ability to realize the above phenomena necessitates the use of multilayer graphene over single-layer graphene. The physics of Fig. 1 is generic to all methods of doping, e.g., electrostatic gating and charge transfer. Employing NLG and α -RuCl₃ charge-transfer heterostructures, we successfully realized all of the phenomena summarized in Fig. 1.

Our discoveries are presented as follows. In Sec. II, we employ second harmonic generation imaging to verify that the stacking-dependent displacement field controls inversion symmetry. We then show that the displacement field mixes the Raman and infrared activity for the optical phonon modes in the inversion-symmetry-broken heterostructures. Crucially, the all-vdW stacking facilitates uninhibited access to the NLG plasmonic response through infrared near-field microscopy (nano-IR), unlike previous NLG devices whose top surface is obscured by gate electrodes. In Sec. III, our combined theoretical and experimental analysis of plasmon propagation reveals that the multiple valence bands of NLG cross the Fermi level. We show that the wavelength of the Dirac multiband plasmon becomes insensitive to displacement fields upon doping into all valence bands. Meanwhile, Sec. IV presents more dramatic changes to the plasmon propagation by sandwiching a thin α -RuCl₃ slab between two NLG sheets (NLG/R/NLG). We discover split plasmonic bands as a result of plasmon-plasmon coupling, permitting extraordinary field confinements at ultrahigh carrier densities. Similar to previous investigations of charge transfer in vdW heterostructures [12,15], our findings are consistent across several samples. We conclude that multilayered heterostructures composed of NLG and α -RuCl₃ reveal novel properties of room-temperature collective modes in NLG.

II. INVERSION-SYMMETRY-CONTROLLED OPTICAL PHONONS

We begin our investigation of collective modes in NLG under strong displacement fields and carrier densities by examining collective lattice vibrations known as optical phonons. The ability to excite a phonon directly with light is governed by the symmetry of the host crystal—typically an immutable property in bulk compounds. However, Fig. 2(a) illustrates how displacement fields generated across NLG/ α -RuCl₃ interfaces necessarily break inversion symmetry. (The weaker symmetry-breaking effect of the substrate is ignored.) We probe this symmetry-breaking effect with an established optical probe of inversion symmetry: second-harmonic generation (SHG; see Appendix B). Previously, an emergent SHG signal has been detected in BLG as a result of a weaker charge-transfer compound [16]. Here, we detect strong SHG emission from inversion-symmetry-broken NLG/ α -RuCl₃

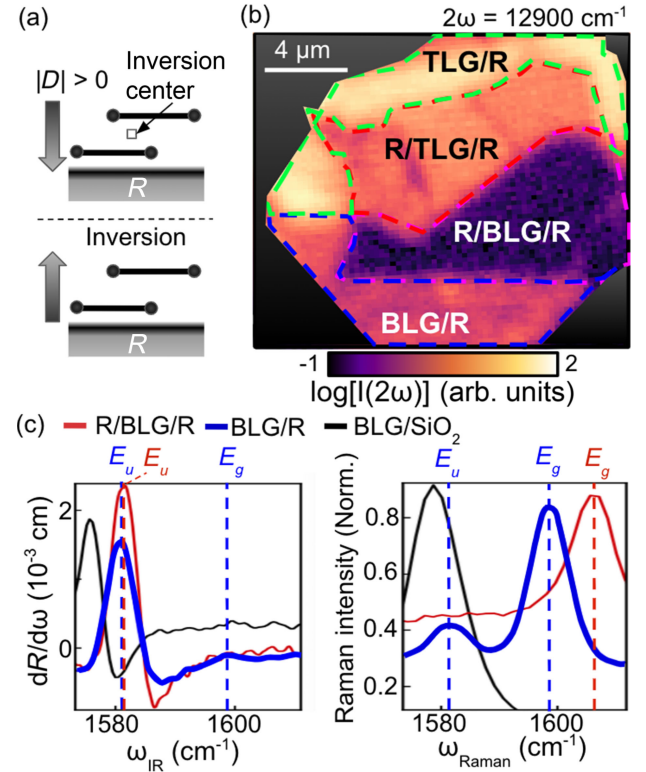


FIG. 2. Controlling optical phonon selection rules in BLG with inversion symmetry. Panel (a) demonstrates how a strong displacement field D breaks inversion symmetry in BLG/R. The weak symmetry-breaking effect of the substrate is ignored. In (b), we display a far-field SHG image of a heterostructure labeled with the four main regions examined in the text. Grayed-out areas block regions with irrelevant layers (see Appendix A for details). Panel (c) shows infrared (left) and Raman (right) spectra of the BLG phonon resonances for three unique heterostructures. Labels E_u and E_g denote phonons with out-of-phase and in-phase interlayer atomic displacements, respectively.

heterostructures. We consider a stack containing four distinct regions of bilayer and trilayer graphene: BLG/R, TLG/R, R/BLG/R, and R/TLG/R (Appendix A describes the stacking procedure). Since the charge transfer is localized to the NLG/R interface [12], the thickness of α -RuCl₃ does not affect the carrier density and displacement field in NLG. We also do not consider the flipped heterostructure R/NLG, as it yields the same net carrier density and displacement field as NLG/R, and our bulk probes are insensitive to the direction of the carrier density distribution in NLG. We deliver a laser pulse of duration ~ 100 fs centered at $\omega = 6450$ cm⁻¹ to the sample and plot the SHG intensity map in Fig. 2(b). Emission intensity at 2ω is highly sensitive to the stacking configuration. The contrast between BLG/R and R/BLG/R is consistent with inversion symmetry breaking in BLG/R and inversion symmetry in R/BLG/R.

Because of the natural broken inversion symmetry in ABA-stacked trilayer graphene [17], TLG regions exhibit higher SHG intensity than BLG. The TLG/R SHG intensity is stronger than that of R/TLG/R, confirming previous observations that displacement fields enhance existing optical nonlinearities in NLG [16]. In Appendix B, we argue that absorption from the top α -RuCl₃ layer in R/NLG/R regions is insufficient to explain the reduction of SHG strength relative to NLG/R regions. Appendix B, Fig. 7 also confirms that emission spectra are dominated by SHG signals. In contrast, up-converted nonlinear photoluminescence obscures SHG characterization of exfoliated NLG flakes [16,18], suggesting that such signals are quenched by the charge transfer from α -RuCl₃. Thus, Fig. 2(b) demonstrates that multi-interface heterostructures of NLG and α -RuCl₃ exhibit controllable inversion symmetry.

Next, Fig. 2(c) shows how inversion symmetry affects the BLG optical phonon selection rules. The left-hand panel displays infrared differential reflectance data $dR/d\omega$ (see Appendix C) for three separate BLG heterostructures: R/BLG/R, BLG/R, and BLG/SiO₂. In all three spectra, a resonance can be observed near $\omega_{\text{IR}} \sim 1580$ cm⁻¹. The line shape of this antisymmetric mode [E_u , Fig. 1(c)], evolves with doping in accordance with the “phonon anomaly” discussed at length elsewhere [2,3,5]. The BLG/R infrared spectrum also displays a small peak at 1599 cm⁻¹, unlike R/BLG/R and BLG/SiO₂ spectra. A secondary phonon resonance has previously been inferred from spectral weight analysis of BLG infrared reflectance [19], but has not been directly observed. However, observation of a second phonon feature is commonplace in Raman spectroscopy and used as evidence for inversion symmetry breaking [1,20]. Therefore, we compare our infrared data with Raman spectra for the same three samples [right-hand panel of Fig. 2(c)]. The Raman spectra exhibit greater peak position variation among the three samples, reflecting the stronger doping sensitivity of the E_g phonon [20]. The BLG/R spectrum indeed displays two modes, while BLG/SiO₂ and R/BLG/R spectra reveal only one resonance. In Appendix C, Fig. 8, we map the

double Raman resonances over a large BLG/R sample area. The relative uniformity of the double resonance feature rules out doping inhomogeneity as a possible explanation. Crucially, the peak energies of the two BLG/R resonances, labeled E_u and E_g , are identical between Raman and infrared measurements (highlighted by the blue dashed lines in both panels), while the relative intensities are reversed. Finally, we remark that none of these phonon resonances are observed on as-exfoliated α -RuCl₃ flakes. Thus, the inversion symmetry breaking in BLG/R mixes optical phonon selection rules, prescribing a small IR activity to the E_g phonon of BLG.

III. DIRAC MULTIBAND PLASMONS

Next, we consider SPP propagation in NLG and α -RuCl₃ heterostructures. Owing to the robustness of our theoretical model relating the SPP wavelength to the carrier density and displacement field (described below), this section offers a more quantitative understanding of the charge-transfer characteristics than the optical phonon measurements presented in the previous section. To date, SPP propagation in vdW charge-transfer heterostructures have been limited to studies of single-layer graphene. Here, we will show that, like SLG, plasmons become immune to displacement fields upon doping into all NLG valence bands. SPP imaging and interferometry rely on the subdiffractive imaging capabilities of infrared near-field optical microscopy (nano-IR; see Appendix D) [6,12,14]. The metallized tip of an atomic force microscope is illuminated with midinfrared light [Fig. 3(a)] that is then elastically backscattered to a detector. We limited our illumination frequency range to 850 – 1000 cm⁻¹, avoiding propagation losses from phonon resonances in the SiO₂ substrate and h -BN superstrate (see Fig. 9 of Appendix D for a simulation of the plasmon dispersion over a more extended frequency range). A map of the local plasmonic field in the underlying heterostructure is then obtained as the tip raster scans across the surface. The nano-IR light probes layers up to ~ 10 nm below the top surface [21,22], affording greater flexibility in the placement of target layers within vertical heterostructures compared to more surface-sensitive probes (e.g., scanning tunneling microscopy). Thus, SPPs are readily accessible in NLG and α -RuCl₃ heterostructures by virtue of our charge-transfer-based device architecture and the unique capabilities of the nano-IR probe. Upon combining these nano-IR results with our modeling, we establish record-high carrier densities and displacement fields among charge-transfer heterostructures.

Figure 3 overviews the main results of plasmon propagation in heterostructures composed of NLG and RuCl₃. Example raw data files relevant for Figs. 2 and 3 can be found in Ref. [23]. In Fig. 3(b), we consider the same sample as imaged with SHG in Fig. 2(b). We observe that for $\omega = 900$ cm⁻¹, the TLG region surrounded by two

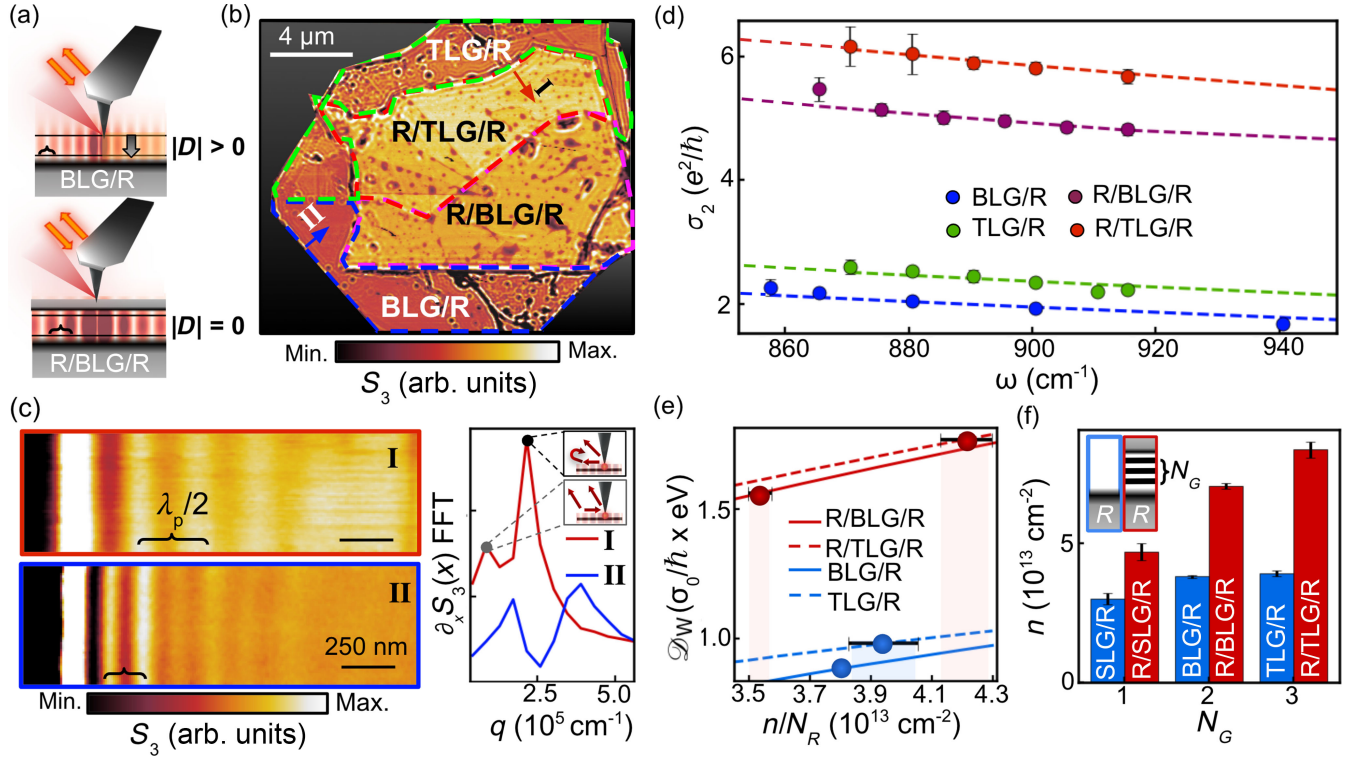


FIG. 3. Overview of plasmon propagation in NLG/R and R/NLG/R heterostructures. Panel (a) sketches plasmon propagation in BLG/R and R/BLG/R heterostructures, where the former exhibits a finite displacement field D . In each case, a midinfrared laser is incident on the tip, which scans over the top surface to map out the SPP propagation with wavelength λ_p . In (b), we display a nano-IR overview image ($\omega = 900 \text{ cm}^{-1}$) of the same terraced heterostructure as in Fig. 2(b). The red and blue arrows follow SPP propagation in the R/TLG/R (I) and BLG/R (II) regions, respectively. The left-hand panel in (c) displays the enlarged plasmon profiles I and II ($\omega = 915 \text{ cm}^{-1}$). Fast Fourier transforms (FFT) of the profiles are displayed on the right-hand panel, and illustrations of the tip- and edge-launched-plasmon interference schemes are provided in the inset. In panel (d), we plot the imaginary part of the optical conductivity determined from the plasmon wave vector [Eq. (E1)]. The fits (dashed lines) enable direct determination of the Drude weight $\mathcal{D}_W$. Then panel (e) converts the Drude weight to carrier density n via a theoretical calculation of the Drude weight as a function of carrier density n/N_R , where N_R is the number of $\alpha\text{-RuCl}_3$ interfaces constructed with methodology described in the main text. The measured Drude weights are denoted by the data points, with the corresponding carrier density on the horizontal axis. Panel (f) summarizes the carrier densities for all samples fabricated in this work, including for single-layer graphene.

RuCl_3 interfaces (R/TLG/R) exhibits the strongest near-field signal S , while the BLG/R region displays the weakest response. Despite the obvious inhomogeneous carrier distribution over large length scales within the stack, nano-IR imaging seamlessly identifies clean areas that support unobstructed SPPs. Example clean areas are highlighted by arrows I and II in the R/TLG/R and BLG/R regions, respectively. Enlarged nano-IR profiles, aligned with I and II, are displayed in Fig. 3(c), revealing periodic, exponentially decaying profiles characteristic of SPP propagation. In general, nano-IR imaging of SPPs exhibits more than one spatial periodicity [24]. For example, the closest fringe spacing, termed “tip-launched” plasmons, displays a periodicity of $\lambda_p/2$ as a result of interference between tip-scattered laser light and plasmons that launch from the tip, reflect from a nearby edge, and scatter again from the tip. In contrast, the weaker and longer-period “edge-launched” plasmons exhibit a wavelength of λ_p ,

interferences that arise between edge-scattered laser light and plasmons that launch from the edge and scatter from the tip. We quantitatively confirm the presence of these two periodicities with a fast-Fourier transformation (FFT) of both line profiles I and II [right-hand panel of Fig. 3(c)]. By comparing the FFT peak positions of the stronger tip-launched mode at $q = 4\pi/\lambda_p$, we conclude that profile I possesses a significantly longer plasmon wavelength λ_p compared to profile II. From the relation $\lambda_p \propto \mathcal{D}_W/\omega^2$, we thus infer a larger Drude weight in region I. Therefore, the data in Figs. 3(b) and 3(c) suggest that regions with the most graphene layers and $\alpha\text{-RuCl}_3$ interfaces display the longest plasmon wavelengths and strongest midinfrared near-field signals. We observe a more modest nano-IR contrast between R/TLG/R and R/BLG/R regions in Fig. 3(b) than the corresponding SHG contrast in Fig. 2(b), suggesting that small doping variations are not required to account for their nonlinear optical response.

These trends are representative of many samples fabricated for each layer arrangement.

By employing standard fitting procedures [12,24], the profiles in Fig. 3(c) access the complex plasmon wave vector $q(\omega) = 2\pi/\lambda_p + i/L_p$, where L_p is the plasmon propagation length. In Appendix E, we show that $q(\omega)$, derived from the SPP line profile, directly determines the complex optical conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$ for each of the regions labeled in Fig. 3(b). In Fig. 3(d), we plot $\sigma_2(\omega)$, extracted from plasmon wave vectors at several IR frequencies ω . For quantitative accuracy, we combine data from multiple samples, selecting clean plasmon profiles such as Fig. 3(c), free of inhomogeneity. The error bars originate from the confidence interval of fits to the real-space SPP line profiles. The evident inverse relationship $\sigma_2 \propto 1/\omega$ suggests that fitting the data to a Drude model (dashed lines) is appropriate:

$$\sigma(\omega) = \frac{i\mathcal{D}_W}{\omega + i\gamma}. \quad (2)$$

Appendix A shows that the Drude weight parameter $\mathcal{D}_W$ varies by approximately $\sim 10\%$ across five samples of the same stacking arrangement, consistent with previously measured doping variations in SLG/R heterostructures [12,15].

Next, we extract the carrier density n from the Drude weight $\mathcal{D}_W$. We employ a tight-binding + Hartree calculation, described in Appendixes G and I, to plot the Drude weight versus carrier density n/N_R in Fig. 3(e) (solid and dashed curves). The overlaid data points denote the Drude weights obtained from Fig. 3(d), enabling conversion to the carrier density on the horizontal axis. The carrier densities, determined through this combined experimental-theoretical approach, are summarized in Fig. 3(f). Our results for SLG are referenced on the leftmost columns, where the scaling $\mathcal{D}_W^{(\text{SLG})} \propto |E_F|$ is well established. Blue bars denote single-sided NLG/R heterostructures, while red bars denote double-sided R/NLG/R heterostructures. Both stacking arrangements reveal increasing carrier densities with additional graphene layers N_G . At fixed N_G , R/NLG/R heterostructures intuitively exhibit larger carrier densities than their NLG/R counterparts due to the presence of an additional charge-transfer interface. Note that the carrier density for R/SLG/R is below twice the SLG/R carrier density, a consistent result understood by considering the increasing work function in SLG with hole doping. This discrepancy appears to reduce for R/TLG/R, as the carriers may distribute across the additional layers without increasing the work function. In Appendix F, we consider the scattering rate parameter γ , discovering a systematic decrease with the number of graphene layers N_G , while increasing with the number of α -RuCl₃ interfaces N_R .

The carrier densities in Fig. 3(f) straightforwardly determine the displacement field at the charge-transfer interface: $D_{\text{NLG/R}} = en$, $D_{\text{R/NLG/R}} = en/2$ [Eq. (I8)]. Previous studies of bilayer graphene on CrCl₃ obtained interface

displacement fields up to 2.5 V/nm [13]. Here, we achieve displacement fields nearly 3 times this value, providing a quantitative underpinning for the level of control of the optical phonons reported in the previous section. However, the interface displacement field is nonzero for R/NLG/R heterostructures because the field is not evaluated at the inversion center of the heterostructure. Thus, the average displacement field $\langle D \rangle$ over all NLG layers [Eq. (I9)] is a more relevant figure of merit for the inversion symmetry breaking demonstrated in the previous section. This latter quantity requires calculation of the screened carrier density profile n_ℓ in NLG/R and R/NLG/R (Appendix I). Figure 4(a) shows the result of such a calculation for TLG. For TLG/R,

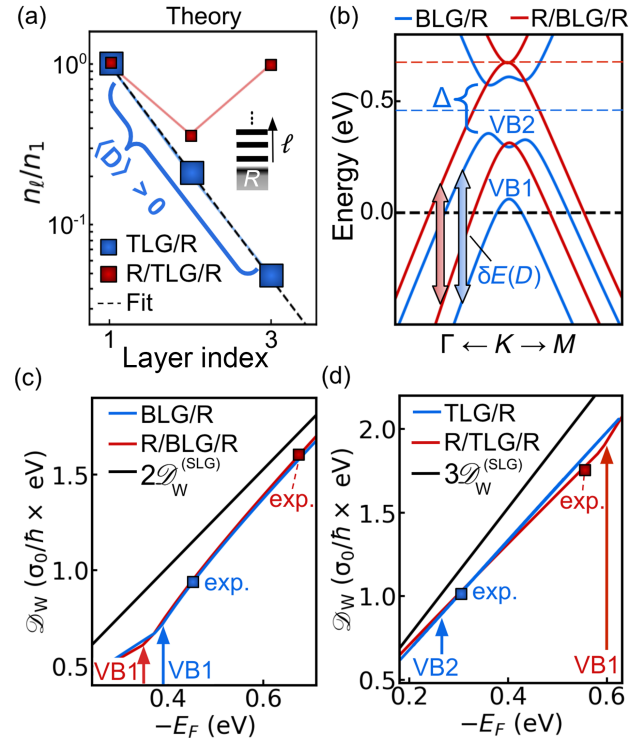


FIG. 4. Dirac multiband plasmons in NLG and α -RuCl₃ heterostructures. The calculated screened carrier density profile n_ℓ/n_1 in TLG/R and R/TLG/R is presented in (a). The layer-averaged displacement field is nonzero only in TLG/R. The energy bands of BLG/R (blue) and R/BLG/R (red) heterostructures are displayed in (b). The BLG/R energy gap $\Delta = 240$ meV is labeled, along with the band splitting $\delta E(D)$ between valence bands 1 and 2 (denoted VB1 and VB2, respectively). The colored dashed lines denote the midpoint between valence and conduction bands, and the black dashed line indicates the Fermi level. The Fermi energy dependence of the Drude weight, derived from these band structures, is then shown in (c). The experimentally obtained Fermi energies (data points) are larger than the onset energy of the inner band VB1 (arrows). The BLG/R and R/BLG/R Drude weights are bounded below the black solid line, denoting the Drude weight from two decoupled graphene layers, $\mathcal{D}_W^{(\text{SLG})}$. The Fermi energy dependence of the TLG Drude weight is displayed in (d), where the onset of the bands VB2 and VB1 are labeled for TLG/R and R/TLG/R, respectively.

TABLE I. Summary of total carrier densities n , interface displacement fields $D_{\text{interface}}$, and layer-averaged displacement fields $\langle D \rangle$ [Eq. (19)] obtained in this work. Parentheses denote the uncertainty on the trailing digit.

Sample	n (10^{13} cm^{-2})	$D_{\text{interface}}$ (V/nm)	$\langle D \rangle$ (V/nm)
SLG/R	3.0(2)	5.4(3)	...
R/SLG/R	4.7(3)	4.2(3)	...
BLG/R	3.8(1)	6.9(2)	4.0(2)
R/BLG/R	7.1(2)	6.4(2)	0
TLG/R	3.9(1)	7.0(2)	2.8(2)
R/TLG/R	8.4(4)	7.6(4)	0

the $e^{-(\ell-1)/L}$ fit (black dashed line) indicates that the carrier density n_ℓ/n_1 decays over the length $L = 0.65$ layers away from the charge-transfer interface and results in a finite average displacement field. On the other hand, the carriers are distributed symmetrically in R/TLG/R, since the charge transfer from each α -RuCl₃ layer into TLG can be assumed to be identical. The resulting displacement field between layers 1 and 2 is equal and opposite to that between layers 2 and 3, resulting in $\langle D \rangle = 0$. The influence of the displacement field gradient $D_{23} - D_{12}$ on the collective modes is a higher-order effect and requires further study. With the help of Eq. (18), Table I summarizes the carrier densities and displacement fields obtained in this work. Of all vdW charge-transfer heterostructures considered to date, NLG/R heterostructures produce the largest displacement fields, while R/NLG/R heterostructures yield the highest carrier densities.

The sizable internal displacement field $\langle D \rangle$ results in significant changes to the NLG band structure, such as the opening of a large band gap. At energies well outside of the gap, the displacement field has the effect of increasing the band energy separation to the amount δE . In Appendix J, we show that δE increases with displacement field via the simple expression

$$\delta E(D) = \sqrt{4t_1^2 + \Delta(D)^2}, \quad (3)$$

where t_1 is the interlayer tight-binding parameter (Appendix G), and Δ is the band gap. The BLG/R and R/BLG/R bands are displayed in Fig. 4(b), where the band splitting between valence bands 1 and 2 (VB1 and VB2, respectively) is labeled. TLG band structures are found in Appendix I. In these calculations, hybridization with RuCl₃ bands is neglected [25], as the relevant energy scales therein are considerably smaller than room temperature. In BLG/R, we calculate a $\Delta = 240$ meV band gap, on par with infrared spectroscopy measurements of dual-gated BLG devices operating at maximum displacement fields [26]. Meanwhile, the Fermi energy (dashed line) is situated at $E_F = -430$ meV below the band gap midpoint, where band curvature is minimal. In R/BLG/R, we observe that

bands remain ungapped, consistent with the preserved symmetry of 180° rotations about the armchair axis of the central layer. Likewise, the Fermi energy is shifted further to $E_F = -650$ meV below the band edge, with the VB2-VB1 band splitting identical to that of bare BLG: $\delta E(0) = 2t_1$. In both BLG/R and R/BLG/R situations, the Fermi level is placed within both valence bands. We will verify shortly that plasmonic effects under displacement fields are nullified upon doping into all NLG bands, as achieved for BLG/R heterostructures. In TLG/R heterostructures, only the outer two bands VB3 and VB2 are depleted (Appendix G, Fig. 12), and thus displacement fields are more relevant.

To consider how the plasmon evolves with Fermi energy and displacement field, Fig. 4(c) plots the Drude weight versus Fermi energy for R/BLG/R ($\langle D \rangle = 0$) and BLG/R ($\langle D \rangle > 0$). Consistent with the increased band splitting illustrated in Fig. 4(b), the primary effect of the displacement field is to slightly delay the kink in the Drude weight, corresponding to the onset of VB1 (arrows). The experimentally obtained Fermi energies (data points) are above this kink, confirming the multiband nature of the plasmon. At the highest Fermi energies, $-E_F \rightarrow \infty$, the Drude weights of both BLG/R and R/BLG/R curves converge toward that of two decoupled monolayers, $2\mathcal{D}_W^{(\text{SLG})} = 8\sigma_0|E_F|/\pi$, denoted by the solid black line. This convergence is understood in the context of Eq. (1) by considering the BLG bands $E_\beta(k) \sim \frac{1}{2}[-\sqrt{(2\hbar v_F k)^2 + t_1^2} + (-1)^\beta t_1]$ in the $k \rightarrow \infty$ limit, where the Dirac dispersion yields $S_\beta v_\beta \rightarrow |E_F| + (-1)^\beta \delta E/2$ for valence bands $\beta = 1, 2$. Therefore, the BLG plasmonic response is well described by a Dirac multiband plasmon and approaches the displacement-field-independent limit $\mathcal{D}_W \sim 2|E_F|$.

Contrary to BLG/R plasmons, a modest displacement field effect is expected for TLG/R plasmons. Since carriers deplete only the outer two valence bands (Appendix I, Fig. 12), the plasmon wavelength $\lambda_p \sim \sum_{\beta=2,3} \mathcal{D}_W^{(\beta)}$ remains sensitive to the band shift $\delta E_\beta(D)$, such that $\mathcal{D}_W^{(\beta)} \sim 4\sigma_0[|E_F| + \delta E_\beta]/\pi$. In Fig. 4(d), we display the Drude weight as a function of Fermi energy for TLG/R (blue line), R/TLG/R (red line), and three decoupled graphene layers (black line). Upon doping into VB2, the TLG/R Drude weight overtakes that of R/TLG/R (for R/TLG/R, VB2 is ungapped). As a result, the measured R/TLG/R Drude weight at $E_F = -0.55$ eV is nearly 10% smaller than the prediction for TLG/R at the same Fermi energy. In Appendix J, we verify that $\sum_{\beta=1,2,3} \delta E_\beta = 0$, and thus the TLG/R and R/TLG/R Drude weights only converge toward the decoupled $3\mathcal{D}_W^{(\text{SLG})}$ limit upon doping into innermost band VB1 near $E_F \approx -0.6$ eV (red arrow). We conclude from Fig. 4 that, in NLG/R heterostructures, few-layer graphene is required to alter Dirac multiband plasmons with displacement fields.

IV. ENHANCED CONFINEMENT FROM INTERLAYER PLASMONIC COUPLING

So far, we have investigated the properties of collective modes in NLG and α -RuCl₃ heterostructures under strong displacement fields and carrier densities. While phonons are susceptible to inversion-symmetry-breaking displacement fields, Dirac multiband plasmons are unperturbed in the $-E_F \rightarrow \infty$ limit. On the other hand, the symmetry of the highly doped NLG plasmons can be altered upon introducing interlayer plasmonic coupling. Specifically, the confinement of plasmons can be engineered by controlling the hybridization of two plasmonic sheets separated by a α -RuCl₃ slab of thickness d_R . In this situation, the α -RuCl₃ serves two purposes. First, it provides charge carriers for the NLG sheets at each NLG/R interface. Because of the interfacial nature of the charge transfer, the carrier density in each NLG sheet is equivalent to that summarized in Table I. Additionally, the α -RuCl₃ serves as a dielectric spacer, where mode coupling between the NLG layers splits the wave vector q of a single plasmonic sheet into two branches [10,27,28], q_S and q_A . This mode splitting is highly sensitive to the α -RuCl₃ thickness, unlike

the charge-transfer carrier density. The low-momentum mode S is symmetric in its in-plane electric field $E_x(x, z)$, while the high-momentum mode A is antisymmetric about the vertical midpoint of the α -RuCl₃ slab [Fig. 5(a)]. In NLG/R/NLG heterostructures, the plasmon coupling coefficient [10] $g \sim e^{-qd_R}$ becomes the control parameter, rather than the displacement field $\langle D \rangle$ described in the previous section.

Experimental investigations of coupled graphene plasmons are scarce due to the challenging fabrication requirements of multigated samples. Here, the vdW charge-transfer platform considerably reduces the device complexity. The antisymmetric plasmon is a crucial ingredient for the charge-transfer SPP tool kit, enabling systematic study of SPPs in the regime of both strong field confinement and high carrier densities. Owing to its reduced damping γ (Appendix F, Fig. 10), bilayer graphene is chosen as the host medium, sandwiching RuCl₃ on both sides (BLG/R/BLG). A α -RuCl₃ spacer thickness of 44 nm is chosen to tune the symmetric and antisymmetric modes within our accessible momentum window.

Using nano-IR imaging, Fig. 5(b) displays real-space profiles $S_4(x)$ of in-plane plasmon propagation for BLG/R

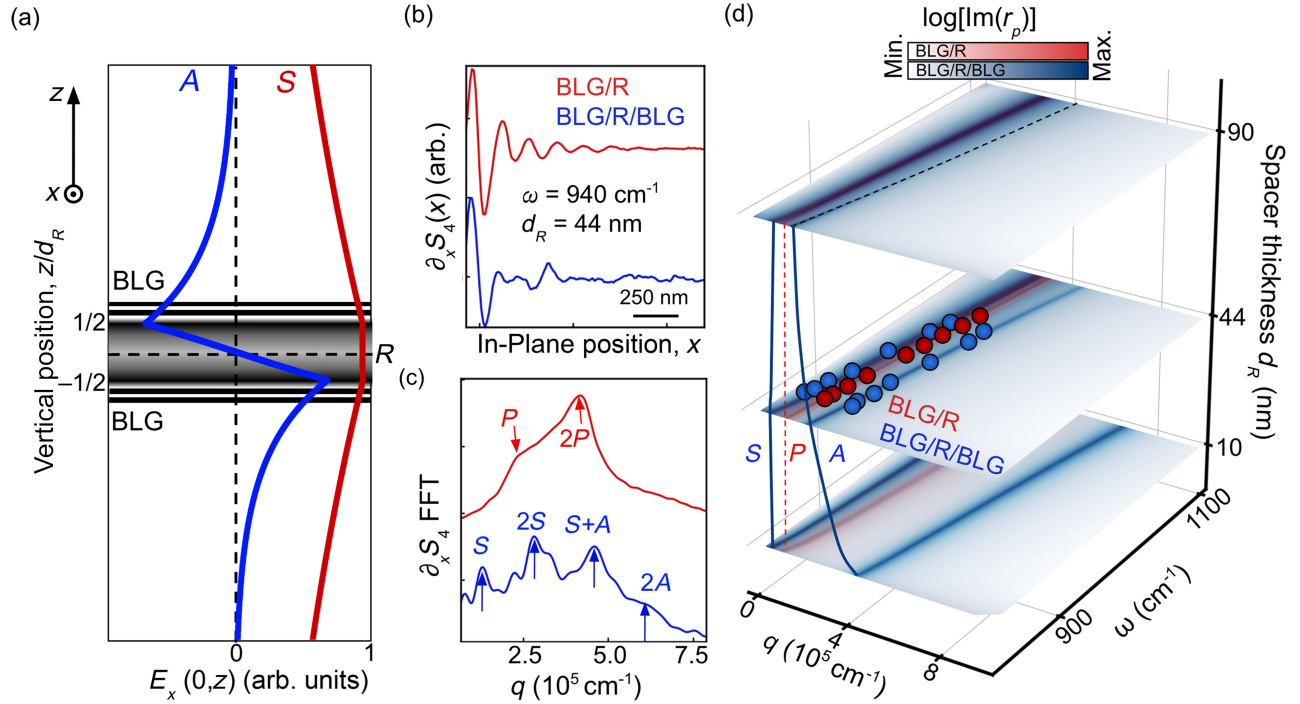


FIG. 5. Interlayer plasmon-plasmon coupling in NLG/R/NLG heterostructures. In (a), we compute the electric field profile $E_x(x=0, z)$ of the symmetric (red) and antisymmetric (blue) plasmons, superimposed on a schematic of the NLR/R/NLG heterostructure. Here, $z=0$ denotes the vertical midpoint of the RuCl₃ slab of thickness d_R . Panel (b) shows example nano-IR profiles $\partial_x S_4(x)$ for BLG/R (red) and BLG/R/BLG (blue) heterostructures at infrared frequency $\omega = 940 \text{ cm}^{-1}$ and RuCl₃ thickness $d_R = 44 \text{ nm}$. Panel (c) displays the corresponding Fourier transform, where the peaks (arrows) are assigned to linear combinations of wave vectors q_S and q_A (while the notation P is reserved for the uncoupled BLG/R plasmon). Panel (d) displays the p -polarized reflection coefficient versus frequency ω , momentum q , and thickness d_R . For clarity, a black dashed line traces the antisymmetric mode at $d_R = 90 \text{ nm}$. The fitting-extracted wave vectors of BLG/R (red) and BLG/R/BLG (blue) are overlaid onto the simulated p -polarized reflectivity for $d_R = 44 \text{ nm}$ (data points).

(red) and BLG/R/BLG (blue) heterostructures at $\omega = 940 \text{ cm}^{-1}$. Slowly varying backgrounds were removed with the spatial derivative, $\partial_x S_4(x)$. Figures 13(a) and 13(b) of Appendix K provide a large-area nano-IR image of the sample used to measure these line profiles. There, the single-sided BLG/R region exhibits the weakest nano-IR signal, consistent with the imaging results of Fig. 3(b). To understand the intricate oscillations of the BLG/R/BLG data, we perform a Fourier transformation in Fig. 5(c). We reveal four momentum peaks for BLG/R/BLG (blue) and two peaks for BLG/R (red). The two-peak structure (labeled P) is characteristic of tip- and edge-launched plasmons, as mentioned earlier. On the other hand, the presence of four peaks implies multimodal plasmon propagation. The peaks are identified as combinations of S (for the symmetric mode) and A (for the antisymmetric mode). The strength and momenta of the combination mode $S + A$ displays no dependence on laser power, suggesting a mode coupling effect that remains within the linear optical regime.

Figure 5(d) corroborates the experimental data with a simulation of the coupled plasmon dispersion $q_{S,A}(\omega)$ versus α -RuCl₃ spacer thickness d_R . The 3D plot displays fixed d_R slices of the imaginary component of the p -polarized reflection coefficient vs frequency and momentum, i.e., $\text{Im}[r_p(\omega, q; d_R)]$. The maxima in these color plots correspond to the dispersion $q_{S,A}(\omega)$. The BLG/R plasmon dispersion $q_P(\omega)$ is superimposed for reference. This calculation is highly sensitive to the BLG conductivity (see Appendix E). According to the excellent agreement between the measured plasmon wave vectors and the calculated plasmon dispersion $\text{Im}[r_p]$ in Fig. 5(d), we find that the BLG/R Drude weight obtained in Fig. 3(e) adequately captures the experimental data of this section.

The antisymmetric BLG/R/BLG plasmon dispersion reveals significant dependence on d_R for $d_R < 100 \text{ nm}$. In particular, the A mode shifts from the uncoupled BLG/R wave vector near $d_R = 100 \text{ nm}$ to beyond our maximum near-field momentum sensitivity, 10^6 cm^{-1} , at $d_R = 10 \text{ nm}$. In contrast, the symmetric mode S shifts to a lower momentum. The intermediate thickness fabricated in this work, $d_R = 44 \text{ nm}$, is therefore optimal for the nano-IR detection of the antisymmetric plasmon. We fit the nano-IR profiles to a multimodal model with the same functional form as mentioned in Sec. III [12,24]. After applying this procedure to all measured excitation frequencies ω , we experimentally obtain the plasmon dispersions $q_S(\omega)$, $q_A(\omega)$, and $q_P(\omega)$. The results (data points) are overlaid onto the $d_R = 44 \text{ nm}$ plane. We find that the experimental wave vectors follow the predicted dispersions, thereby validating a gate-free realization of interlayer-coupled plasmons. In Appendix K, we compare this BLG/R/BLG experimental data with that from a thicker α -RuCl₃ spacer, 56 nm , finding that the antisymmetric plasmon wave vector reduces in momentum as predicted by Fig. 5(d). With

charge transfer vdW heterostructures, the interlayer coupling may be programmed with increasing complexity by incorporating additional NLG/RuCl₃ pairs, e.g., BLG/R/BLG/R/BLG.

V. CONCLUSION

We have demonstrated that displacement fields and interlayer coupling are effective tools for controlling collective modes in heterostructures composed of NLG and α -RuCl₃. Notably, these tools are absent in SLG. The displacement field breaks inversion symmetry in NLG/R, as evidenced by strong second-harmonic generation and mixed phonon selection rules. On the other hand, displacement fields are absent in R/NLG/R heterostructures, negating the effects presented in the asymmetric stacks. The 7 V/nm displacement fields and $8 \times 10^{13} \text{ cm}^{-2}$ carrier densities reported here are outmatched only by ion-gel gated devices. We discover a new plasmonic collective mode, the Dirac multiband plasmon, which arises when the Fermi level crosses multiple quasilinear valence bands in NLG. We realize strong plasmon-plasmon coupling in NLG/R/NLG heterostructures, unlocking confined plasmon propagation at ultrahigh carrier densities.

Future studies are needed to examine these collective modes under the regime of low doping and high displacement fields, which may be realized by interfacing NLG and α -RuCl₃ with an n -type charge-transfer compound, such as ZrO_x [29]. Ultimately, this limit could pave the way for topologically protected plasmons at Bernal-stacked domain walls [30,31]. While we have constrained our study to phonons and plasmons at room temperature, the prospect of examining more exotic collective excitations, such as Cooper pairs in superconducting rhombohedral-stacked TLG/R heterostructures [32] and excitons in α -RuCl₃ [33], offer to deepen our understanding of correlated electron systems.

ACKNOWLEDGMENTS

Research at Columbia is supported by DOE-BES-SC0018426. Development of nanoimaging techniques at Columbia is supported as part of Programmable Quantum Materials, an Energy Frontiers Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under Award No. DE-SC0019443. M. S. S. was supported by grant PRE2021-097070 from Ministerio de Ciencia, Innovación y Universidades through Agencia Estatal de Investigación. K. S. B. is grateful for the support of the National Science Foundation, Award No. DMR-2310895.

DATA AVAILABILITY

Data supporting Figs. 2 and 3 are openly available [23], and additional data supporting the findings of this article are available from the authors upon reasonable request.

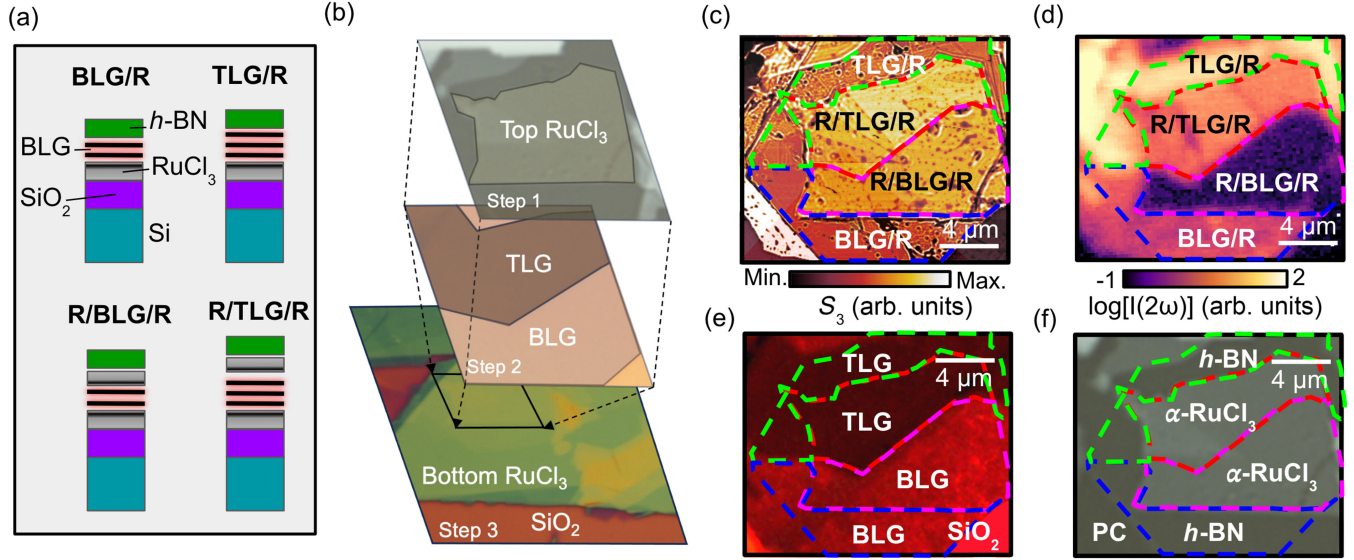


FIG. 6. Comparing experimental imaging data with the constituent layers of the heterostructure. Panel (a) reviews the layer arrangements of the heterostructure presented in Figs. 2(b) and 3(b): BLG/R, R/BLG/R, TLG/R, and R/TLG/R. Next, panel (b) illustrates the assembly steps of the heterostructure shown in Figs. 2(b) and 3(b) using the dry-transfer method, described in Appendix A. The flake borders are outlined and their contrast is artificially enhanced for clarity. In the optical microscope image labeled “step 1” (top panel), the top α -RuCl₃ layer of the heterostructure is picked up beneath the *h*-BN encapsulating layer with PC. In step 2 (middle panel), the terraced NLG flake is picked up below the *h*-BN/RuCl₃ layers. Finally, in step 3, the *h*-BN/RuCl₃/NLG stack is dropped down onto the bottom α -RuCl₃ flake, which rests on an SiO₂/Si substrate. Panels (c) and (d) repeat Figs. 2(b) and 3(b), without grayed-out borders. Panel (d) displays a high-contrast optical microscope image of the NLG flake prior to stacking, with the TLG and BLG regions labeled. The colored boundaries of (b) and (c) are overlaid on top for reference. Panel (e) shows the first two layers of the stack—*h*-BN and α -RuCl₃—on the PC transfer slide, with the same colored outlines overlaid for comparison.

APPENDIX A: HETEROSTRUCTURE ASSEMBLY

Crystal growth details can be found in an earlier work on Graphene/RuCl₃ heterostructures [12]. The standard dry-transfer technique was used, employing polycarbonate (PC) as the stacking polymer. In all stacks described in the main text ~ 5 nm hexagonal boron nitride was chosen as the top layer in order to protect RuCl₃ and NLG from ambient or solvent exposure. In order to reliably pick up thin RuCl₃, we treated SiO₂/Si substrates with 1-dodecanol for 5 min at 160°C prior to exfoliation. To minimize bubble formation in the stacks, which inhibits plasmon readout due to doping inhomogeneity, the stacking was carried out at relatively

high temperature ($\sim 140^\circ\text{C}$) during and after α -RuCl₃ pickup steps. The bottom α -RuCl₃ thickness ranged from 40 to 100 nm, a value that did not affect the charge-transfer density into NLG. Meanwhile, to ensure adequate tip-plasmon coupling, the top thickness ranged from 3 to 10 nm. The completed stack was dropped onto blank SiO₂/Si substrates at 180°C–190°C. The surface was then vacuum annealed overnight at 180°C. These stacking procedures are summarized in Figs. 6(a) and 6(b) for the case of the heterostructure presented in Figs. 2(b) and 3(b). Table II suggests $\sim 10\%$ doping variation across different samples, consistent with previous measurements of SLG/R heterostructures [12,15].

Figures 6(c)–6(f) demonstrate that the constituent layers of the heterostructure presented in Figs. 2(b) and 3(b) align with the artificial dashed colored outlines presented in those images. For example, Fig. 6(e) shows that the border between R/TLG/R and R/BLG/R labels aligns precisely with the terrace between the TLG and BLG regions. Likewise, Fig. 6(f) demonstrates precise alignment at the boundary between $N_R = 1$ and $N_R = 2$ regions, corresponding to the top α -RuCl₃ flake. In these images, we also confirm that grayed-out boundaries correspond to regions where relevant flakes are missing from the stack (e.g., *h*-BN or NLG) and/or extraneous layers (e.g., additional RuCl₃) are present.

TABLE II. Measured Drude weights of five BLG/R and R/BLG/R heterostructures.

Sample	$\mathcal{D}_W$ ($\sigma_0/\hbar \times \text{eV}$)	
	BLG/R	R/BLG/R
1	0.90	1.41
2	1.00	1.56
3	0.87	1.58
4	0.89	1.48
5	0.88	1.58
Average	0.91	1.52
Std. Dev.	0.05	0.07

APPENDIX B: SHG IMAGING SUPPLEMENTARY INFORMATION

Second-harmonic generation measurements were conducted on a modified commercial confocal microscope (Nikon Eclipse Ti), customized for nonlinear optical microscopy. The optical pump was generated by a Coherent Chameleon Ultra II Ti:sapphire oscillator (pulse duration 150 fs and repetition rate 80 MHz) tuned to 865 nm and down-converted to 1550 nm using an optical parametric oscillator (Coherent Chameleon OPO compact-Vis). The pump beam was attenuated to ~ 4.5 mW, spectrally filtered using a 950-nm long-pass filter (Thorlabs), and coupled into the microscope using a 950-nm short-pass dichroic filter. The pump was then focused onto the sample using a $100\times$ Nikon objective with a numerical aperture of 0.95. The SHG signal was collected in a backscattering geometry and filtered by a series of 950-nm short-pass filters. The second harmonic signal was then relayed to either an avalanche photodiode (MPD photonics) for SHG intensity mapping or a grating spectrometer (Princeton instruments) for hyperspectral characterization.

To model the effect of transmission losses from the top RuCl_3 , we consider light incident from vacuum onto a RuCl_3 film of thickness d_{topR} , then exiting into vacuum onto the other side (air/ RuCl_3 /air). Considering a reasonable dielectric function of $\epsilon(2\omega) \approx 10 + 2i$ [33], the transmission through the thin RuCl_3 may be readily computed. The 10% transmission loss through a $d_{\text{topR}} = 5$ nm top RuCl_3 flake is insufficient to explain the order-of-magnitude change in SHG intensity from TLG/R to R/TLG/R.

Figure 7 demonstrates that signals measured in Fig. 2(b) are dominated by SHG, with multiphoton PL backgrounds at least a factor of 10 weaker in strength. We perform a hyperspectral line scan from top to bottom of the image in Fig. 2, covering all TLG/R, R/TLG/R, R/BLG/R, and BLG/R regions. The nonlinear PL background is observed as a broad band from 13000 to 14000 cm^{-1} . This suggests that, upon high doping, nonlinear PL signals are quenched,

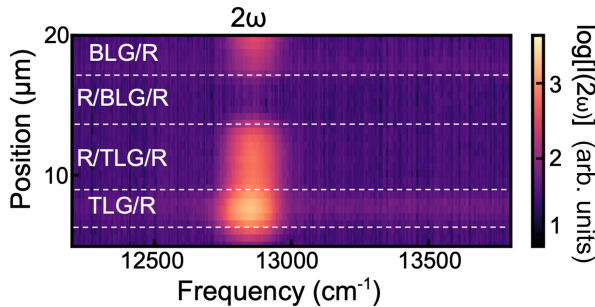


FIG. 7. Hyperspectral SHG line scan. The line scan starts at the top region of Fig. 2(b), corresponding to the TLG/R region, then ends at the BLG/R region in the bottom region of Fig. 2(b). The SHG energy is labeled on the top of the frame by 2ω , where $\omega = 6450 \text{ cm}^{-1}$ is the pump frequency.

while their strength in charge-neutral NLG is of similar order to that of the SHG signal [17]. A minute SHG signal is observed in the R/BLG/R region, which may be tied to weak symmetry-breaking effects of planar interfaces.

APPENDIX C: OPTICAL PHONON SPECTROSCOPY SUPPLEMENTARY INFORMATION

Reflectance measurements (relevant for infrared spectroscopy) were performed with the Bruker Hyperion microscopy tool, using a Global thermal source and a KBr beam splitter. The R/BLG/R and BLG/R stacks were transferred onto an SiO_2/Si chip containing a nearby gold pad, where background measurements were collected.

Raman spectroscopy measurements were performed with a Horiba Micro-Raman tool under 532-nm excitation. Spectra were acquired for 30–60 sec, and the grating resolution was set to 1800 lines/mm.

To verify that the double-peak feature observed in the BLG/R Raman spectrum of Fig. 2 is tied to inversion symmetry breaking, we performed Raman mapping over a large-area BLG/R sample. The results are shown in Fig. 8. An example spectrum in Fig. 8(a) reproduces that observed in Fig. 2(c). The peak intensities and frequencies are denoted $I_{G\pm}$ and $\omega_{G\pm}$, respectively. We then plot $\omega_{G+} - \omega_{G-}$ vs I_{G+}/I_{G-} in Fig. 8(b) for all data points extracted from the Raman intensity map I_{G+}/I_{G-} in Fig. 8(c). The uniformity of the double-peak structure is evident, suggesting that doping inhomogeneity cannot explain our observations.

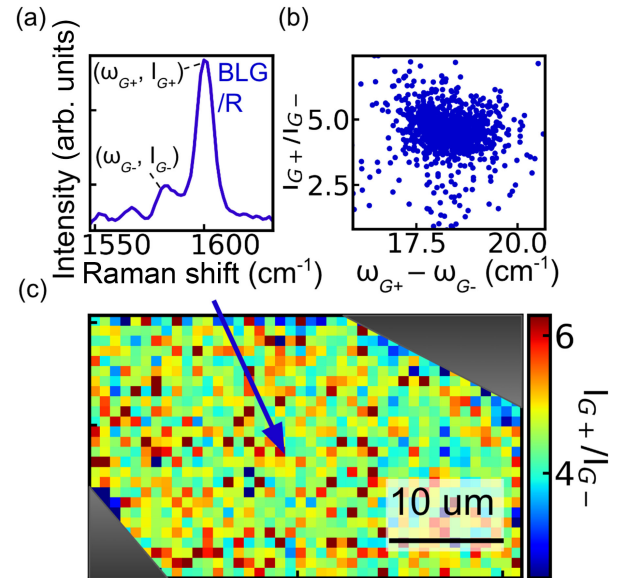


FIG. 8. Mapping of split peak in a large-area BLG/R heterostructure. Panel (a) shows an example spectrum. There, the peak energies and intensities ($I_{G\pm}$, $\omega_{G\pm}$) are labeled. Panel (b) plots the ratio I_{G+}/I_{G-} vs $\omega_{G+} - \omega_{G-}$ for all data points measured in the I_{G+}/I_{G-} map (c).

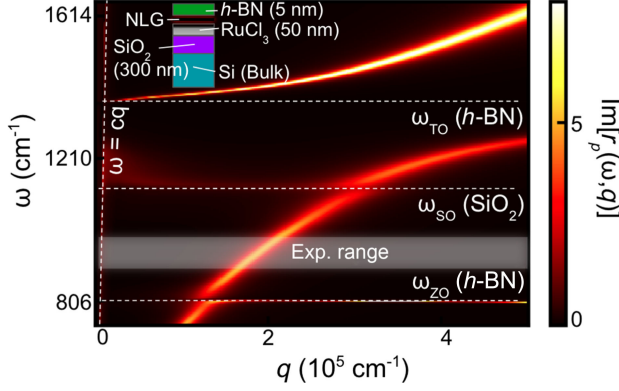


FIG. 9. Simulated NLG/R plasmon dispersion in a realistic heterostructure consisting of 5 nm of top *h*-BN and 300 nm of SiO₂ on bottom. The *h*-BN and SiO₂ phonons, as well as the experimentally interrogated frequency range, are labeled.

APPENDIX D: NANO-IR MEASUREMENT DETAILS

An incident, broadly tunable IR laser with frequency ω is chosen to excite SPP propagation. The laser scatters off of a metallized tip and is sent to a mercury cadmium telluride detector. The tip oscillates at frequency Ω . Upon interferometrically detecting the demodulated signal at harmonics of Ω (via the standard pseudo-heterodyne technique [34]), we readily obtain background-free signals with amplitude and phase information. The data were acquired with a Neaspec GmbH laser (QCL) from Daylight Solutions. A custom PYTHON code was used to analyze the plasmon profiles. We constrained the QCL frequency range to ~ 850 – 1000 cm⁻¹, where plasmons propagate unimpeded by phonon resonances in surrounding layers (see Fig. 9).

APPENDIX E: PLASMON EXTRACTION OF OPTICAL CONDUCTIVITY

Consider a heterostructure with N separate conducting 2D sheets, surrounded by media with thicknesses $\{d\}$ and dielectric functions $\{\epsilon\}$. Then the p -polarized transfer matrix $\mathbf{M}_p(\omega, q)$ can be computed given an in-plane wave vector q . The p -polarized reflection coefficient is then given by the elements $r_p(\omega, q) = M_{10}/M_{00}$. The complex plasmon wave vector q_* is a pole of r_p ; i.e., $M_{00}(\omega, q_*) = 0$.

Here, we wish to determine the conductivity from the plasmon dispersion q_* . In general, we may write

$$M_{00}(\omega, q) = \prod_i [a_i(\omega, q, \{d\}, \{\epsilon\}) + \sigma_i(\omega) b_i(\omega, q, \{d\}, \{\epsilon\})]. \quad (\text{E1})$$

Therefore, $\sigma_i(\omega) = -a_i(\omega, q_*)/b_i(\omega, q_*)$. Note that this relation holds for only one sheet i . This procedure is underdetermined unless there is only one unknown

conducting sheet, or all conducting sheets can be assumed identical.

Coefficients a_i and b_i can be computed with the aid of a symbolic algebraic programming language (here the PYTHON SYMPY package is used). Consider one sheet. Then one calculates $M_{00}(\omega, q_*; \sigma)$ and notes that $a_i = M_{00}(\omega, q_*; 0)$ and $b_i = \partial_\sigma M_{00}(\omega, q_*; 0)$. In this manner, the complex conductivity is fully determined. For an arbitrary number of identical sheets, more derivatives ∂_σ must be computed.

APPENDIX F: SCATTERING RATE ANALYSIS FOR NLG AND RuCl₃ HETEROSTRUCTURES

Figure 10 plots the scattering rate parameter γ , obtained from Eq. (2), for several charge-transfer heterostructures. Red and blue data points show γ vs number of graphene layers N_G for R/NLG/R and NLG/R heterostructures, respectively. We compare these data with scattering rates obtained from a heterostructure composed of graphene and RuCl₃, separated by a variable number of *h*-BN layers N_{BN} (green data points). Note that blue and green data points denote separate SLG/R heterostructures for $N_{BN} = 0$, $N_G = 1$. We find that scattering rates from all datasets exhibit a monotonic decrease with layer numbers. Meanwhile, R/NLG/R stacks exhibit the largest scattering rates, consistent with prior conclusions implicating RuCl₃ as the primary scattering channel [12]. Losses reduce with increasing N_G for R/NLG/R and NLG/R, mimicking the trend of the SLG/BN/R dataset. Since γ is a layer-averaged quantity, the data suggest that dissipation channels predominate at the nearest G-RuCl₃ interface. We conclude that in charge-transfer heterostructures, BLG and TLG plasmons exhibit improved propagation losses compared to monolayer graphene plasmons.

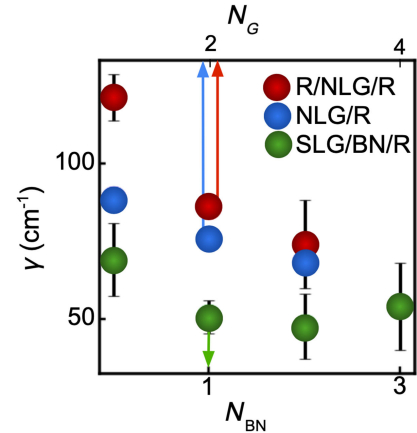


FIG. 10. Scattering rate parameter γ extracted from Drude fitting to plasmon dispersions. The top horizontal axis is the number of graphene layers N_G , relevant for NLG/R and R/NLG/R structures, while the bottom horizontal axis denotes the number of BN spacer layers used in SLG/BN/R stacks.

APPENDIX G: TIGHT-BINDING MODEL

In the NLG/RuCl₃ structures, the hybridization gaps are $\lesssim 3$ meV [35]. Hence, those effects will be washed out in our room-temperature experiments, $k_B T = 25$ meV. Consequently, we make the approximation that the species are coupled only through the dipole field induced by the transfer of charge due to their work function difference, to be explained below.

For the description of the electronic bands of the NLG structures, we employ a tight-binding model with five parameters: t_0, t_1, t_3, t_4 , and Δ' in the notation of Ref. [36]; see Fig. 11. t_0 is the hopping integral between nearest neighbors in the same layer, controlling the Fermi velocity of the Dirac cone, $v_F = 0.3236 \times |t_0(\text{eV})| \times 10^8$ cm/s. t_1 is the vertical hopping between sublattice *A* and *B* atoms on contiguous layers. t_3 is the hopping between atoms from *A* and *B* sublattices on contiguous layers, separated by a horizontal distance equal to the carbon-carbon distance in graphene, a_{cc} . t_4 is the hopping between atoms from the same sublattice on contiguous layers, separated by a horizontal distance of a_{cc} . Reference [36] also includes the additional parameters t_2 and t_5 . Their magnitudes are of the order of several meV and affect only the physics at much smaller densities than those of our interest in this work [37]. Finally, Δ' is an on-site potential for atoms with a vertical neighbor on proximal layers (*B* atoms in the bottom layer and *A* atoms in the top layer in Fig. 11).

We fix $t_0 = -3.1$ eV, corresponding to a Fermi velocity of $v_F = 10^8$ cm/s. For bilayer graphene, we set the remaining parameters according to Ref. [38]: $t_1 = 0.381$ eV, $t_3 = 0.380$ eV, $t_4 = 0.140$ eV, and $\Delta' = 0.022$ eV. For trilayer graphene we use the parameters of Ref. [37]:

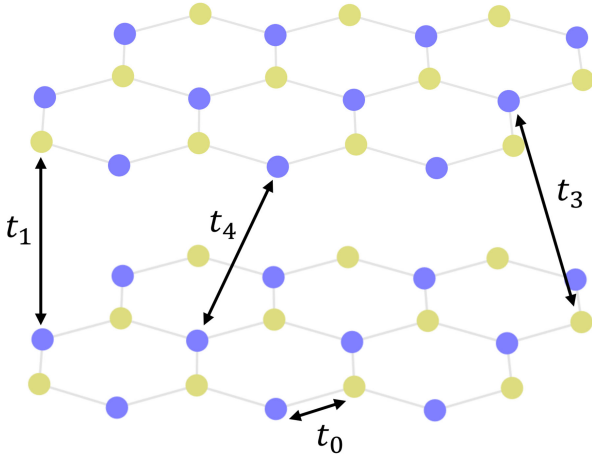


FIG. 11. A bilayer of graphene is depicted in the energetically favorable Bernal, or AB, stacking. Atoms from the *A* sublattice (yellow) in the top layer sit on top of atoms from the *B* sublattice (blue) in the bottom layer. In structures with more layers this basic stacking is repeated. The tight-binding hoppings are also depicted.

$t_0 = -3.1$ eV, $t_1 = 0.380$ eV, $t_3 = 0.290$ eV, $t_4 = 0.141$ eV, and $\Delta' = 0.035$ eV.

Finally, in the Drude weight computations we will make the assumption that it comes entirely from the doped graphene layers, for the flat Hubbard bands of the RuCl₃ will display weak infrared response. The Drude weights are computed at $T = 0$ because their temperature dependence is negligible at finite dopings [39].

APPENDIX H: CONDUCTIVITY AND DRUDE WEIGHT

In layered materials the electrical conductivity is a matrix with layer indices $\ell_{1,2}$, $\sigma_{\ell_1 \ell_2}^{\alpha\beta}(q, \omega)$.

Plasmons are the solutions to a matrix equation involving the conductivity and the photon propagator [40]. In this work, we are interested in the solutions in which the electric field is in phase in all the layers, giving rise to a dispersion $\omega_{pl} \sim \sqrt{q}$ at low momenta [15]. For these modes, up to corrections of order e^{-qd} where d is the interlayer distance, the material behaves as a 2D system with the total electrical conductivity $\sigma^{\alpha\beta}(q, \omega) = \sum_{\ell_1 \ell_2} \sigma_{\ell_1 \ell_2}^{\alpha\beta}(q, \omega)$. Setting $\lambda \sim 200$ nm and $d = 3.3$ Å yields $e^{-qd} \sim 0.99$. In addition, we may take the long wavelength limit $q \rightarrow 0$ relevant for the plasma dispersion [41], $\sigma^{\alpha\beta}(q, \omega) \approx \sigma^{\alpha\beta}(0, \omega) \equiv \sigma^{\alpha\beta}(\omega)$. Finally, time-reversal invariance and sixfold rotations in graphene make the conductivity tensor proportional to the identity, $\sigma^{\alpha\beta}(\omega) = \sigma(\omega)\delta^{\alpha\beta}$.

Neglecting interband processes, the conductivity has the general form [42]

$$\sigma(\omega) = \frac{i\mathcal{D}_W}{\omega + i\gamma}, \quad (\text{H1})$$

where $1/\gamma$ is the transport relaxation time of the carriers and $\mathcal{D}_W$ is called the Drude weight. For frequencies much larger than γ , the conductivity is purely imaginary, $\sigma(\omega) = i\mathcal{D}_W/\omega$, and plasmons are long-lived. The Drude weight can be computed with knowledge of the band structure $\epsilon_n(\mathbf{k})$ (with the chemical potential included by setting the zero point of the energy) [43],

$$\mathcal{D}_W = \frac{e^2}{2\pi^2} \sum_n \int d^2k \frac{\partial}{\partial \epsilon_n(\mathbf{k})} [-n_T(\epsilon_n(\mathbf{k}))] |v_n^x(\mathbf{k})|^2, \quad (\text{H2})$$

where n runs over the electron bands, $n_T(\epsilon) = 1/(1 + e^{\epsilon/k_B T})$ is the Fermi distribution function at temperature T , and $v_n^x(\mathbf{k}) = \partial \epsilon_n(\mathbf{k}) / \partial k_x$ the velocity matrix element. The factor of 2 due to spin degeneracy is already taken into account in this expression. For 3D systems the prefactor should be $e^2/(4\pi^3)$.

APPENDIX I: HARTREE THEORY OF SCREENING

To properly account for the charge screening in the charge-transfer structure, we will include the electronic effects in the tight-binding model via the Hartree approximation.

First, let us consider the effect of the dielectric environment on the interaction strength. We model the experimental device as a collection of slabs of dielectrics $i = 1, \dots, N$ with relative permittivities ϵ_{ri} and thicknesses d_i . The graphene layers will be placed in a vacuum gap among the dielectrics, in contact with the bulk RuCl_3 on one side for the one-sided structures and on both sides for the two-sided structures.

After a straightforward calculation, the electrostatic potential in the vacuum gap is

$$U(z, z_0) = -\frac{n_0 e}{2\epsilon_0} \frac{2\epsilon_{r1}}{\epsilon_{r1} + \epsilon_{rN}} |z - z_0| \quad \text{for } z > z_0 \quad (\text{I1})$$

$$= -\frac{n_0 e}{2\epsilon_0} \frac{2\epsilon_{rN}}{\epsilon_{r1} + \epsilon_{rN}} |z - z_0| \quad \text{for } z < z_0, \quad (\text{I2})$$

with n_0 the homogeneous 2D density in the charged layer, $e > 0$ is the electron charge, z is the vertical coordinate where we probe the potential, and z_0 is the position of the charged layer. If the dielectric constants ϵ_{r1} and ϵ_{rN} have frequency dependence, they should be evaluated at $\omega = 0$. The Coulomb Hamiltonian is then

$$H_C = \frac{1}{2} \sum_{\ell \ell'} V(z_\ell, z_{\ell'}) n_\ell n_{\ell'}, \quad (\text{I3})$$

with ℓ, ℓ' denoting the layers and the propagator $V(z_\ell, z_{\ell'})$ is defined by $V(z_\ell, z_{\ell'}) \equiv U(z_\ell, z_{\ell'})/n_{\ell'}$. We can replace $V(z_\ell, z_{\ell'})$ by a symmetrized propagator $[V(z_\ell, z_{\ell'}) + V(z_{\ell'}, z_\ell)]/2$ without changing the Hamiltonian, resulting in the new expression $V(z, z') = -[e^2/(2\epsilon_0)]|z - z'|$. The symmetric propagator is equal to the free vacuum propagator, and the interaction does not depend on the dielectric environment of the experiment. On the other hand, the polarizability of the carbon orbitals in graphene induces an internal effective dielectric constant $\bar{\epsilon} = 1.43$ [44], that should be included. The propagator then reads,

$$V(z, z') = -\frac{e^2}{2\bar{\epsilon}\epsilon_0} |z - z'|. \quad (\text{I4})$$

In the Hartree approximation, the two-body Hamiltonian H_C is decoupled as

$$\begin{aligned} H_C &\approx \frac{1}{2} \sum_{\ell \ell'} V(z_\ell, z_{\ell'}) (\langle n_\ell \rangle n_{\ell'} + n_\ell \langle n_{\ell'} \rangle - \langle n_\ell \rangle \langle n_{\ell'} \rangle) \\ &= \sum_{\ell \ell'} V(z_\ell, z_{\ell'}) \langle n_\ell \rangle n_{\ell'} + \text{const} \\ &\equiv \sum_{\ell} U_\ell n_\ell + \text{const}, \end{aligned} \quad (\text{I5})$$

where $\langle n_\ell \rangle$ denotes the expectation value of n_ℓ .

In addition, assuming a fixed charge-transfer density n at the RuCl_3 that is concentrated on the proximal layers to the graphene, the NLG system will feel an external potential $V_{\text{ext}}(z) = -[(ne^2)/(2\epsilon_z\epsilon_0)]|z - z_c|$, with $\epsilon_z = 2.5$ [44] and z_c the center of charge in the RuCl_3 (a change of the center of charge will only shift the potential on the NLG by a constant independent of z).

The self-consistency process starts by solving the tight-binding model for NLG plus the external potential. By charge conservation, the total density in the NLG will be $-n$ for one-sided structures and $-2n$ for double-sided structures, and the chemical potential is chosen to fix the total density. The density in layer ℓ is obtained straightforwardly:

$$\langle n_\ell \rangle = 2 \left(\frac{1}{A_G N_k} \sum_{\epsilon(n, \mathbf{k}) < 0} \sum_{\alpha} |\psi(n, \mathbf{k})_{\alpha\ell}|^2 \right) - \frac{2}{A_G}. \quad (\text{I6})$$

A_G is the area of the unit cell of graphene and N_k the number of $\mathbf{k}$ points. The sum is over states below the Fermi level, $\epsilon(n, \mathbf{k}) < 0$, and over each atom α in the unit cell belonging to layer ℓ . The factor of 2 comes from spin degeneracy, and we are computing the density relative to the neutral graphene layer with 2 electrons per unit cell.

The typical charge transfer in RuCl_3 -graphene heterostructures is of the order of 10^{13} cm^{-2} , while a filled band corresponds to a density of $A_G^{-1} = 1.9 \times 10^{15} \text{ cm}^{-2}$. To correctly capture the small Fermi pockets without the need for a very fine grid in k space, we consider a patch around the K point with an area of $\lambda^2 = 1/36$ times the area of the Brillouin zone, and cast the grid with N_k points inside this patch. With this setup, the density in layer ℓ is computed as

$$\langle n_\ell \rangle = 4 \left(\frac{\lambda^2}{A_G N_k} \sum_{\epsilon(n, \mathbf{k}) < 0} \sum_{\alpha} |\psi(n, \mathbf{k})_{\alpha\ell}|^2 \right) - \frac{4\lambda^2}{A_G}, \quad (\text{I7})$$

where the factor of 4 comes from the Fermi pocket around the K' point (the two “valleys” are related by time-reversal symmetry) and the spin degeneracy.

Afterward, we compute the Hartree potential of Eq. (I5) and solve the tight-binding model plus the external and Hartree potentials. We use a damping parameter, adjusted at each iteration, that mixes the new and initial densities for better convergence. The newly obtained densities are fed again into Eq. (I5) and the process is repeated. Self-

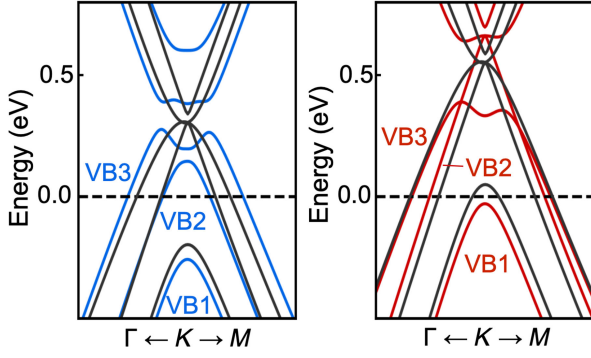


FIG. 12. Band structure for TLG/R (left) and R/TLG/R (right). Gray bands are reference calculations of charge-neutral TLG under zero displacement field, shifted to match the Fermi energy of either TLG/R or R/TLG/R.

consistency is reached when the sum over the layers of the absolute value of the difference between the electrostatic potentials from consecutive iterations, before applying the damping, is less than 1 meV. That is, we stop when $\sum_{\ell} |\delta U_{\ell}| < 1$ meV, with δU_{ℓ} the difference between the potentials from consecutive iterations before the damping.

The screened carrier density profile derived in this appendix triggers the formation of internal displacement fields. The displacement field profile $D_{\ell, \ell+1}$ can be computed between any successive layers $\ell, \ell+1$ via the formula

$$D_{\ell, \ell+1} \equiv \frac{e}{2} (n_{\ell>} - n_{\ell<}), \quad (18)$$

where we use the notation $n_{\ell>} \equiv -n(1 - 1/N_R) + \sum_{\ell'=\ell+1}^{N_G} n_{\ell'}$ and $n_{\ell<} \equiv -n/N_R + \sum_{\ell'=1}^{\ell} n_{\ell'}$. The notation $N_R = (1, 2)$ refers to NLG/R and R/NLG/R heterostructures, respectively, and $n = \sum_{\ell=1}^{N_G} n_{\ell}$ is the net carrier density. In other words, the displacement field between layers ℓ and $\ell+1$ depends on the net carrier densities above and below layer ℓ . We may use definition Eq. (18) to arrive at the results of Table I. First, we consider the interface displacement field, $D_{\text{interface}}$. At the bottom charge-transfer interface, we have $n_{\ell>} = n/N_R$ and $n_{\ell<} = -n/N_R$. Therefore, $D_{\text{interface}} = en/N_R$. We next compute the layer-averaged displacement field:

$$\langle D \rangle = \frac{1}{N_G} \sum_{\ell=1}^{N_G} D_{\ell, \ell+1}. \quad (19)$$

By symmetry $\langle D \rangle = 0$ for R/NLG/R. We therefore consider only single-sided structures ($N_R = 1$). Imposing the carrier density profile ansatz $n_{\ell} = n_1 e^{-(\ell-1)/L}$, we obtain

$$D_{\ell, \ell+1} = en \left(\frac{e^{-\ell/L} - e^{-N_G/L}}{1 - e^{-N_G/L}} \right), \quad (110)$$

from which the average displacement field can readily be computed in terms of the fitting parameter $L = 0.65$ layers [Fig. 4(a)].

In the main text, we showed how displacement fields affect the BLG/R band structure. In Fig. 12, we provide energy bands for TLG/R and R/TLG/R heterostructures, obtained from the tight-binding+Hartree calculations. As expected, TLG/R bands show a distinct energy gap. Despite the expected result $\langle D \rangle = 0$, the R/TLG/R bands are also modified compared to bare TLG bands (dark gray). Here, outer bands are gapped, while middle bands are ungapped and shifted upward. Thus, Fig. 12 hints that internal displacement field gradients are necessary to accurately predict collective mode properties. Most relevant for the Drude response, the innermost bands of R/TLG/R and TLG/R are not yet filled. Based on arguments presented in the next appendix and the main text, this causes deviations between red and blue curves in Fig. 4(d).

APPENDIX J: ANALYTICAL RELATIONSHIP BETWEEN DRUDE WEIGHT AND DISPLACEMENT FIELD

We begin with Eq. (1) under the highly doped regime. In this case, the NLG bands can be considered a sequence of shifted linear bands, each with Fermi velocity v_F . In the space of the valence bands, the projected electronic Hamiltonian in bilayer graphene reads

$$H_{\text{VB}}(k) = \begin{pmatrix} \epsilon^+(k) - E_F & \Delta/2 \\ \Delta/2 & \epsilon^-(k) - E_F \end{pmatrix}, \quad (J1)$$

where $\epsilon^{\pm}(k)$ is the dispersion of the bare inner (outer) valence band, $E_F = -(U_1 + U_2)/2$ and $\Delta = U_1 - U_2$. $U_{1,2}$ are the self-consistently calculated electrostatic potentials on the layers, and the bare energies satisfy $\epsilon^+(k) - \epsilon^-(k) = 2t_1$. We have neglected small contributions coming from the hopping elements t_3 and t_4 and a small k -dependent correction of the off-diagonal element. This approximation is not valid at the K point because the first valence and conduction band become degenerate and should be included in the projected Hamiltonian. However, we are interested in the regime where the Fermi surface of the outer band sits far from the K point. Hence, the effect of the net displacement field is to shift the outer or inner band by $\delta E_{\beta} = \pm(\sqrt{t_1^2 + \Delta^2/4} - t_1)$. We obtain the same result for a calculation incorporating all 4 bands. Hence, our approximation is valid in the high-density regime. Note that the first-order correction to the bands in Δ/t_1 is zero, making band renormalization weak at these high carrier densities.

On the other hand, for trilayer graphene we have

$$H_{\text{VB}}(k) = \begin{pmatrix} \epsilon^t(k) - E_F + \Delta_2 & \Delta_1/\sqrt{2} & -\Delta_1/\sqrt{2} \\ \Delta_1/\sqrt{2} & \epsilon^+(k) - E_F - \Delta_2/2 & 3\Delta_2/2 \\ -\Delta_1/\sqrt{2} & 3\Delta_2/2 & \epsilon^-(k) - E_F - \Delta_2/2 \end{pmatrix}. \quad (\text{J2})$$

$\epsilon^t(k)$ is the monolayerlike band of trilayer graphene and $\epsilon^{+(-)}(k)$ the outer (inner) bilayerlike band. In this case, $E_F = -(U_1 + U_2 + U_3)/3$, $\Delta_1 = (U_1 - U_3)/2$, and $\Delta_2 = (U_1 - 2U_2 + U_3)/6$. In the double-sided structures inversion symmetry is preserved and $\Delta_1 = 0$, whereas in the single-sided structures Δ_1 is parametrically larger than Δ_2 .

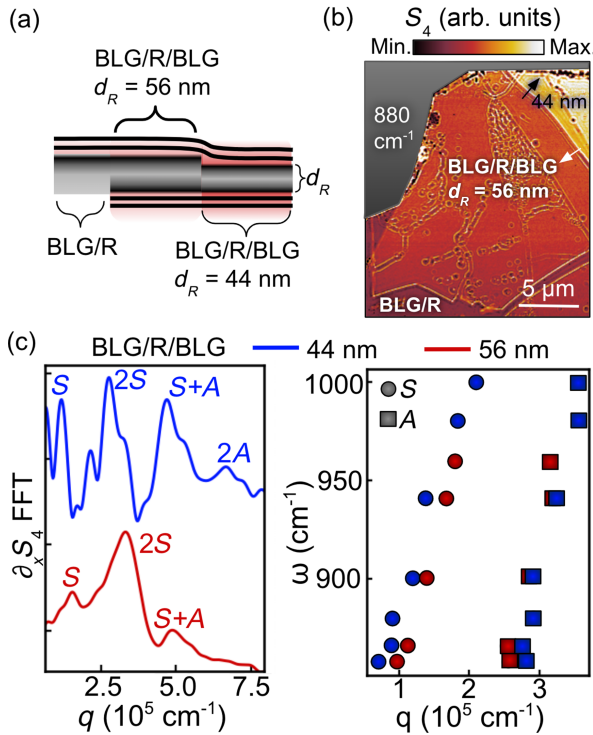


FIG. 13. Extended data for BLG/R/BLG coupled plasmon propagation. Panel (a) shows an overview of the heterostructure, containing regions of both BLG/R and BLG/R/BLG. The α -RuCl₃ thickness is either 56 or 44 nm. Panel (b) shows a large-area nano-IR overview image of this heterostructure, where the three measured regions are labeled (BLG/R; BLG/R/BLG, $d_R = 56$ nm; 44 nm). The gray area blocks off regions that do not contain an encapsulating top h -BN layer. The arrows indicate line profiles used in (c). BLG/R/BLG coupled plasmon propagation for two different α -RuCl₃ spacer thicknesses. In both panels, blue data represent plasmon data from a $d_R = 44$ nm spacer [copied from Figs. 5(b) and 5(c)]. Meanwhile, red data represent data from a thicker, $d_R = 56$ nm, spacer. The left-hand panel shows an FFT of plasmon line profiles at 940 cm⁻¹. The assigned peaks are labeled in the same manner as Fig. 5(d). The right-hand panel shows the two symmetric (circles) and antisymmetric (squares) wave vectors versus incident frequency.

For $\Delta_1 = 0$ the shifts can be obtained analytically: The Dirac band is shifted by $-E_F + \Delta_2$, the outer bilayerlike band by $-E_F - \Delta_2/2 + \sqrt{2t_1^2 + 9\Delta_2^2/4} - \sqrt{2}t_1$, and the inner band by $-E_F - \Delta_2/2 - \sqrt{2t_1^2 + 9\Delta_2^2/4} + \sqrt{2}t_1$. Note that upon doping into all three valence bands, these band shift terms cancel from the net Drude weight. In the experimental R/TLG/R and TLG/R situations, only the outer two valence bands are doped. Thus, the R/TLG/R and TLG/R Drude weight is sensitive to the displacement field, unlike the BLG heterostructures.

APPENDIX K: ADDITIONAL EXPERIMENTAL DATA FOR BLG/R/BLG PLASMON PROPAGATION

We now compare BLG/R/BLG plasmon propagation at two different α -RuCl₃ thicknesses. Figures 13(a) and 13(b) provide a large-area nano-IR overview image of the BLG/R/BLG heterostructure used to interrogate coupled plasmon propagation. As observed previously in Fig. 3(b), the BLG/R region exhibits the weakest nano-IR response as a result of probing the fewest net carriers. Thus, the nano-IR signal increases in the BLG/R/BLG region, and further intensifies as the bottom BLG layer approaches the nano-IR probe—explaining the contrast between BLG/R/BLG regions with $d_R = 44$ nm and $d_R = 55$ nm. The qualitative nano-IR behavior in each region closely mirrors that of Fig. 3, consistent with high doping in each BLG sheet.

Next, the left-hand panel of the Fig. 13(c) shows the FFT of plasmon line profiles for α -RuCl₃ spacer thicknesses of 44 nm [same as Fig. 5(c)] and 56 nm, fixing the frequency to 940 cm⁻¹. The comparison of the coupled plasmon dispersions (right-hand panel) experimentally confirms the prediction from Fig. 5(d): As the thickness increases, the momentum of the symmetric mode increases, while the momentum of the antisymmetric mode decreases. By comparing two different α -RuCl₃ thicknesses, Fig. 13 explicitly highlights the role of α -RuCl₃ as a dielectric spacer in coupled plasmon interactions.

- [1] J. Yan, T. Villarsen, E. A. Henriksen, P. Kim, and A. Pinczuk, *Optical phonon mixing in bilayer graphene with a broken inversion symmetry*, *Phys. Rev. B* **80**, 241417(R) (2009).
- [2] J. Jahng, S. Lee, S. G. Hong, C. J. Lee, S. G. Menabde, M. S. Jang, D. H. Kim, J. Son, and E. S. Lee, *Characterizing*

- and controlling infrared phonon anomaly of bilayer graphene in optical-electrical force nanoscopy, *Light* **12**, 281 (2023).
- [3] J. A. Yan, K. Varga, and M. Y. Chou, *Optical phonon anomaly in Bernal stacked bilayer graphene with ultrahigh carrier densities*, *Phys. Rev. B* **86**, 035409 (2012).
 - [4] F. J. Bezares, A. D. Sanctis, J. R. Saavedra, A. Woessner, P. Alonso-González, I. Amenabar, J. Chen, T. H. Bointon, S. Dai, M. M. Fogler, D. N. Basov, R. Hillenbrand, M. F. Craciun, F. J. García De Abajo, S. Russo, and F. H. Koppens, *Intrinsic plasmon-phonon interactions in highly doped graphene: A near-field imaging study*, *Nano Lett.* **17**, 5908 (2017).
 - [5] A. B. Kuzmenko, L. Benfatto, E. Cappelluti, I. Crassee, D. Van Der Marel, P. Blake, K. S. Novoselov, and A. K. Geim, *Gate tunable infrared phonon anomalies in bilayer graphene*, *Phys. Rev. Lett.* **103**, 116804 (2009).
 - [6] Z. Fei, E. G. Iwinski, G. X. Ni, L. M. Zhang, W. Bao, A. S. Rodin, Y. Lee, M. Wagner, M. K. Liu, S. Dai, M. D. Goldflam, M. Thiemens, F. Keilmann, C. N. Lau, A. H. Castro-Neto, M. M. Fogler, and D. N. Basov, *Tunneling plasmonics in bilayer graphene*, *Nano Lett.* **15**, 4973 (2015).
 - [7] Y. Luan, J. Qian, M. Kim, K. M. Ho, Y. Shi, Y. Li, C. Z. Wang, M. C. Tringides, and Z. Fei, *Imaging stacking-dependent surface plasmon polaritons in trilayer graphene*, *Phys. Rev. Appl.* **18**, 024052 (2022).
 - [8] T. Stauber, P. San-Jose, and L. Brey, *Optical conductivity, Drude weight and plasmons in twisted graphene bilayers*, *New J. Phys.* **15**, 113050 (2013).
 - [9] I. H. Lee, L. Martin-Moreno, P. Avouris, T. Low, and S. H. Oh, *Switching the symmetry of graphene plasmons with nanoemitters for ultimate infrared-light confinement*, *Phys. Rev. Appl.* **19**, 064039 (2023).
 - [10] C. Hu, A. Deng, P. Shen, X. Luo, X. Zhou, T. Wu, X. Huang, Y. Dong, K. Watanabe, T. Taniguchi, G. Xie, X. Li, Q. Liang, and Z. Shi, *Direct imaging of interlayer-coupled symmetric and antisymmetric plasmon modes in graphene/hBN/graphene heterostructures*, *Nanoscale* **13**, 14628 (2021).
 - [11] A. C. Gadelha *et al.*, *Localization of lattice dynamics in low-angle twisted bilayer graphene*, *Nature (London)* **590**, 405 (2021).
 - [12] D. J. Rizzo, B. S. Jessen, Z. Sun, F. L. Ruta, J. Zhang, J. Q. Yan, L. Xian, A. S. McLeod, M. E. Berkowitz, K. Watanabe, T. Taniguchi, S. E. Nagler, D. G. Mandrus, A. Rubio, M. M. Fogler, A. J. Millis, J. C. Hone, C. R. Dean, and D. N. Basov, *Charge-transfer plasmon polaritons at Graphene/ α -RuCl₃ interfaces*, *Nano Lett.* **20**, 8438 (2020).
 - [13] G. Tenasini, D. Soler-Delgado, Z. Wang, F. Yao, D. Dumcenco, E. Giannini, K. Watanabe, T. Taniguchi, C. Mouldsdales, A. Garcia-Ruiz, V. I. Fal'ko, I. Gutiérrez-Lezama, and A. F. Morpurgo, *Band gap opening in bilayer Graphene – CrCl₃/CrBr₃/CrI₃ van der Waals interfaces*, *Nano Lett.* **22**, 6760 (2022).
 - [14] Z. Fei, A. S. Rodin, G. O. Andreev, W. Bao, A. S. McLeod, M. Wagner, L. M. Zhang, Z. Zhao, M. Thiemens, G. Dominguez, M. M. Fogler, A. H. Castro Neto, C. N. Lau, F. Keilmann, and D. N. Basov, *Gate-tuning of graphene plasmons revealed by infrared nano-imaging*, *Nature (London)* **487**, 82 (2012).
 - [15] Y. Wang *et al.*, *Modulation doping via a two-dimensional atomic crystalline acceptor*, *Nano Lett.* **20**, 8446 (2020).
 - [16] M. Zhang, N. Han, J. Wang, Z. Zhang, K. Liu, Z. Sun, J. Zhao, and X. Gan, *Strong second harmonic generation from bilayer graphene with symmetry breaking by redox-governed charge doping*, *Nano Lett.* **22**, 4287 (2022).
 - [17] Y. Shan, Y. Li, D. Huang, Q. Tong, W. Yao, W. T. Liu, and S. Wu, *Stacking symmetry governed second harmonic generation in graphene trilayers*, *Sci. Adv.* **4**, eaat0074 (2018).
 - [18] W. T. Liu, S. W. Wu, P. J. Schuck, M. Salmeron, Y. R. Shen, and F. Wang, *Nonlinear broadband photoluminescence of graphene induced by femtosecond laser irradiation*, *Phys. Rev. B* **82**, 081408 (2010).
 - [19] E. Cappelluti, L. Benfatto, and A. B. Kuzmenko, *Infrared phonon activity and Fano interference in multilayer graphenes*, *Phys. Scr.* **T162**, 014018 (2014).
 - [20] D. L. Mafra, P. Gava, L. M. Malar, R. S. Borges, G. G. Silva, J. A. Leon, F. Plentz, F. Mauri, and M. A. Pimenta, *Characterizing intrinsic charges in top gated bilayer graphene device by Raman spectroscopy*, *Carbon* **50**, 3435 (2012).
 - [21] Y. Dong, L. Xiong, I. Y. Phinney, Z. Sun, R. Jing, A. S. McLeod, S. Zhang, S. Liu, F. L. Ruta, H. Gao, Z. Dong, R. Pan, J. H. Edgar, P. Jarillo-Herrero, L. S. Levitov, A. J. Millis, M. M. Fogler, D. A. Bandurin, and D. N. Basov, *Fizeau drag in graphene plasmonics*, *Nature (London)* **594**, 513 (2021).
 - [22] G. X. Ni, A. S. McLeod, Z. Sun, L. Wang, L. Xiong, K. W. Post, S. S. Sunku, B. Y. Jiang, J. Hone, C. R. Dean, M. M. Fogler, and D. N. Basov, *Fundamental limits to graphene plasmonics*, *Nature (London)* **557**, 530 (2018).
 - [23] S. Moore, *Raw data for Figs. 2 and 3*, figshare.com/projects/nlg-rucl3/248669, 2025.
 - [24] A. Woessner, M. B. Lundberg, Y. Gao, A. Principi, P. Alonso-González, M. Carrega, K. Watanabe, T. Taniguchi, G. Vignale, M. Polini, J. Hone, R. Hillenbrand, and F. H. Koppens, *Highly confined low-loss plasmons in graphene–boron nitride heterostructures*, *Nat. Mater.* **14**, 421 (2014).
 - [25] S. Kim, J. Hong, K. Watanabe, T. Taniguchi, J. Falson, J. Kim, and Y. Kim, *Spin and valley-polarized multiple Fermi surfaces of α -RuCl₃/bilayer graphene heterostructure*, *Appl. Phys. Lett.* **123**, 173101 (2023).
 - [26] Y. Zhang, T. T. Tang, C. Girit, Z. Hao, M. C. Martin, A. Zettl, M. F. Crommie, Y. R. Shen, and F. Wang, *Direct observation of a widely tunable bandgap in bilayer graphene*, *Nature (London)* **459**, 820 (2009).
 - [27] E. N. Economou, *Surface plasmons in thin films*, *Phys. Rev.* **182**, 539 (1969).
 - [28] B. Wang, X. Zhang, X. Yuan, and J. Teng, *Optical coupling of surface plasmons between graphene sheets*, *Appl. Phys. Lett.* **100**, 131111 (2012).
 - [29] B. S. Kim *et al.*, *Ambipolar charge-transfer graphene plasmonic cavities*, *Nat. Mater.* **22**, 838 (2023).
 - [30] E. H. Hasdeo and J. C. Song, *Long-lived domain wall plasmons in gapped bilayer graphene*, *Nano Lett.* **17**, 7252 (2017).

- [31] L. J. Yin, H. Jiang, J. B. Qiao, and L. He, *Direct imaging of topological edge states at a bilayer graphene domain wall*, *Nat. Commun.* **7**, 11760 (2016).
- [32] E. Viñas Boström, A. Fischer, J. B. Profe, J. Zhang, D. M. Kennes, and A. Rubio, *Phonon-mediated unconventional superconductivity in Rhombohedral stacked multilayer graphene*, *npj Comput. Mater.* **10**, 163 (2024).
- [33] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. V. Shankar, Y. F. Hu, K. S. Burch, H. Y. Kee, and Y. J. Kim, *α -RuCl₃: A spin-orbit assisted Mott insulator on a honeycomb lattice*, *Phys. Rev. B* **90**, 041112(R) (2014).
- [34] N. Ocelic, A. Huber, and R. Hillenbrand, *Pseudoheterodyne detection for background-free near-field spectroscopy*, *Appl. Phys. Lett.* **89** (2006).
- [35] J. Shi and A. H. MacDonald, *Magnetic states of graphene proximitized Kitaev materials*, *Phys. Rev. B* **108**, 064401 (2023).
- [36] J. Jung and A. H. MacDonald, *Accurate tight-binding models for the π bands of bilayer graphene*, *Phys. Rev. B* **89**, 035405 (2014).
- [37] A. A. Zibrov, P. Rao, C. Kometter, E. M. Spanton, J. I. A. Li, C. R. Dean, T. Taniguchi, K. Watanabe, M. Serbyn, and A. F. Young, *Emergent Dirac gullies and gully-symmetry-breaking quantum Hall states in ABA trilayer graphene*, *Phys. Rev. Lett.* **121**, 167601 (2018).
- [38] A. B. Kuzmenko, I. Crassee, D. van der Marel, P. Blake, and K. S. Novoselov, *Determination of the gate-tunable band gap and tight-binding parameters in bilayer graphene using infrared spectroscopy*, *Phys. Rev. B* **80**, 165406 (2009).
- [39] T. Stauber, N. M. R. Peres, and A. K. Geim, *Optical conductivity of graphene in the visible region of the spectrum*, *Phys. Rev. B* **78**, 085432 (2008).
- [40] T. Stauber and G. Gómez-Santos, *Plasmons in layered structures including graphene*, *New J. Phys.* **14**, 105018 (2012).
- [41] B. Wunsch, T. Stauber, F. Sols, and F. Guinea, *Dynamical polarization of graphene at finite doping*, *New J. Phys.* **8**, 318 (2006).
- [42] P. Coleman, *Electron transport theory, Introduction to Many-Body Physics* (Cambridge University Press, Cambridge, England, 2015), pp. 332–356.
- [43] N. W. Ashcroft and N. Mermin, *Solid State Physics* (Brooks/Cole, Florence, KY, 1976).
- [44] S. Slizovskiy, A. Garcia-Ruiz, A. I. Berdyugin, N. Xin, T. Taniguchi, K. Watanabe, A. K. Geim, N. D. Drummond, and V. I. Fal’ko, *Out-of-plane dielectric susceptibility of graphene in twistrionic and Bernal bilayers*, *Nano Lett.* **21**, 6678 (2021).

Correction: A funding statement was missing from the Acknowledgments and has been inserted.