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Ultrastrong plasmon-phonon coupling in double-layer graphene intercalated with a transition-metal dichalcogenide

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We pursue the premise that plasmon-phonon coupling and hybrid plasmon-phonon modes can be broadly tailored in van der Waals (vdW) heterostructures. While the coupling between optical plasmons in graphene and phonons of substrate materials has already been widely investigated, the coupling of acoustic plasmons to phonons has remained elusive to date. Here we demonstrate that double-layer graphene intercalated with a transition-metal dichalcogenide (TMD) can harbor *acoustic* plasmon-phonon resonances with particularly high coupling strength. Using the quantum-electrostatic heterostructure method, which takes into account the contribution of each vdW monolayer at the *ab initio* level, we present the dependence of the plasmon-phonon coupling strength on the thickness of the TMD, as well as on the graphene doping. Our results reveal optimal and experimentally feasible conditions to achieve ultrastrong plasmon-phonon coupling, and thus enable further advances in nanoscale thermal and optical devices of high sensitivity.

I. INTRODUCTION

The collective excitations of the two-dimensional (2D) electron liquid in graphene, a 2D honeycomb lattice of carbon atoms [1, 2], are known as Dirac plasmons [3]. In the past several years, graphene plasmonics has attracted pertinently increasing research attention, following the discoveries of strong electromagnetic field confinement [3, 4], characteristic frequencies tunable by the Fermi energy [5–8], and particularly low losses [9, 10]. Relevant to a broad range of optical devices, it was shown that graphene can support plasmons with frequencies from mid-infrared [11–13] to terahertz range [7, 13–15]. Furthermore, graphene can be combined with other 2D materials, such as MX₂ transition metal dichalcogenides (TMDs, e.g. MoS₂ and WS₂), with a metal (M) layer between two layers of a chalcogen (X) atoms, enabling versatile van der Waals heterostructures (vdWhs) [16–18]. It is nowadays possible to precisely control not only constituents but also their precise stacking in a vdWhs [16, 19–24], as well as arrange and adjoin them laterally rather than on top of one another [23–29]. As a result, one may confidently claim the tailor-made multi-layered artificial vdWhs to bolster specific properties and/or desired behavior [18, 30]. In that sense, particularly strong advances have been made in optoelectronics [16–19, 21, 23–31], and graphene plasmonics has emerged as a promising platform for next-generation optoelectronic nanodevices, such as tunable

photodetectors [32], mechanical tuning absorbers [33], optoelectronic switches [34], optical modulators [35, 36], and sensors [37, 38].

Fig. 1(a, b) illustrates a heterostructure made of N layers of a TMD material sandwiched inside a double-layer graphene (DLG). When this system is excited by light, a hybrid excitation known as surface plasmon-phonon polariton (SPPP) arises. These quasi-particles emerge when phonons in the TMD are coupled to the electron oscillations in graphene [4]. One of the possibilities to experimentally investigate (excite and measure) the properties of this hybrid excitation is to use scatter-type scanning near-field optical microscopy (s-SNOM) [15, 39, 40]. This allows one to measure in the real space the wavelength of SPPPs, with a resolution down to 20 nm [5, 6, 39–44].

Although TMDs with hexagonal symmetry (2H-TMDs) present four non-degenerate optical phonon modes in the infrared (IR) spectrum at the Γ -point, only two of them can couple to Dirac plasmons, via the long-range Fröhlich coupling near the phonon frequencies, namely, the in-plane E' , which is IR and Raman (R) active, and out-of-plane A_2'' (IR) [19, 45–47]. For an even number of stacked TMD layers, the E' and A_2'' modes of 1L-MX₂ split into E_u (IR) and E_g^1 (R), and A_{2u} (IR) and A_{1g}^1 (R) modes, respectively [45]. For bulk MX₂ on the other hand, A_2'' splits into A_{2u} (IR) and B_{2g}^1 modes, where the latter is optically inactive (silent). Fig. 1(c) illustrates the described evolution of the A_2'' mode from a single layer to MX₂ bulk case. For more details and a complete description of N -layer 2H-TMDs phonon modes at the Γ -point, we refer to Refs. 21 and 47.

Recently, graphene-based systems with coupled plasmon-phonon polaritons have been proven useful for

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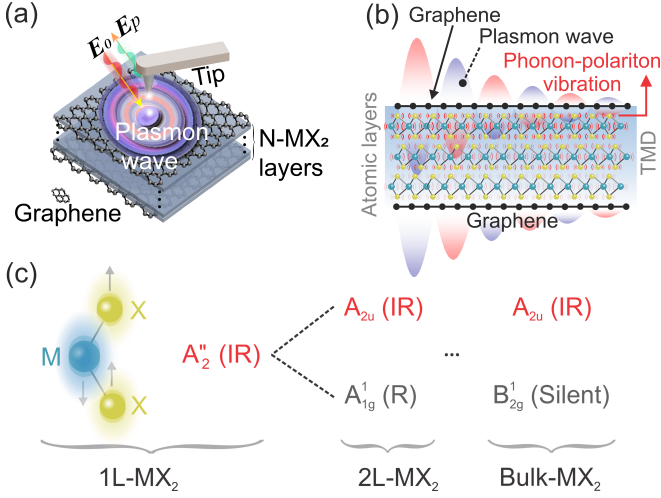


FIG. 1. (Color online) (a) Schematic illustration of the scatter-type scanning near-field optical microscopy setup and a Dirac plasmon wave in van der Waals heterostructure (vdWhs) composed by N -MX₂ ($M = \text{Mo, W}$; $X = \text{S, Se}$) layers encapsulated by two graphene (G) monolayers. (b) Lateral view of the G/3-MX₂/G vdWhs, illustrating the hybridization of TMD phonon vibration with Dirac plasmon in graphene, giving rise to surface plasmon-phonon polaritons (SPPs). (c) Schematic representation of the out-of-plane A₂^{''} phonon mode in a MX₂ monolayer. For an even number of TMD layers the A₂^{''} mode splits into A_{2u}^{''} (IR) and A_{1g}^{''} (R). For the bulk-MX₂, A₂^{''} splits into A_{2u}^{''} (IR) and B_{2g}^{''}, where the latter is silent for excitations. Here IR and R stand for infrared and Raman, respectively.

near-field radiative heat transfer, which may improve the heat dissipation capacity of nanodevices [48–50]. Furthermore, previous works on double-layers have shown that electric fields of the plasmons of the two plasmonic interfaces will couple and split plasmon modes into two, namely the optical and the acoustic mode [51–55]. One of the advantages of using DLG-based vdWhs is that one can have a dual-gate control of the carrier density individually in either graphene layer, and the possibility to control interlayer interactions by introducing another 2D material between the graphene monolayers, yielding uniquely novel electronic and optical properties. More recently, acoustic plasmons were visualized in real-space in large-area graphene, truly opening technological possibilities to use acoustic plasmons in graphene-based optoelectronics and sensing applications [56].

Motivated by above facts, in this paper we have designed a double-layer graphene-based vdWhs, illustrated in Fig. 1(a, b), aimed at enhancing and tuning the coupling of *acoustic* Dirac plasmons of graphene to the out-of-plane IR-active A₂^{''} phonon mode of the TMD inside the DLG. To characterize the desired coupling and the resultant properties, we used realistic simulations at the level of density function theory (DFT) and many-body perturbations, combined with the quantum electrostatic heterostructure (QEH) model. We thereby identify the

way in which the coupling between acoustic Dirac plasmons and out-of-plane IR-active A₂^{''} phonon modes of MoS₂ and WS₂ takes place, and how it depends on the number of TMD layers put between the graphene ones. We further reveal that doping of graphene layers can be used to broadly tune the coupling strength, enabling the versatile design of optoelectronic and thermal nanodevices.

As shown in Ref. 57, the acoustic Dirac plasmon in graphene couples mostly to the out-of-plane IR-active A₂^{''} TMD phonon, hence in this work we maintain the focus on the coupling to that particular mode. We however consider double-layer graphene, in order to generate an inter-layer plasmon-plasmon interaction, thereby increasing the magnitude of the confined perpendicular electric field mode [58–60], and, consequently, further enhancing the acoustic Dirac plasmon coupling to the out-of-plane IR-active A₂^{''} TMD phonon mode.

The paper is organized as follows. In section II, we introduce the random phase approximation theory (RPA) of plasmons in spatially separated double-layer graphene and of hybrid plasmon-phonon polaritons modes. In section III, we present the results of the plasmon-phonon dispersion in the (q, ω) -plane obtained from the QEH model, emphasizing the way we calculate the plasmon-phonon coupling strength (Ω) at the minimum energy splitting (MES). Then, we show the dependence of the SPPs coupling strength on the number of TMD layers, for the acoustic plasmons and IR-active out-of-plane A₂^{''} phonon mode, and identify weak (WC), strong (SC), and ultra-strong (USC) coupling regimes, via the normalized coupling parameter η . Finally we discuss how the doping of graphene and the consequently varied Fermi energy affect the coupling strength. The summary of our findings and conclusions is provided in section IV.

II. THEORY OF PLASMONS IN DOUBLE-LAYER GRAPHENE AND HYBRID PLASMON-PHONON POLARITONS MODES

Dirac plasmons, defined as density oscillations of Dirac-like low energy electrons in graphene, can be obtained from the total dielectric function of the system within the random phase approximation (RPA) [60–62]. For a double-layer graphene system, the collective plasmon modes are obtained by solving the plasmon equation, which corresponds to the zeroes of the generalized dielectric tensor $\epsilon_{ij}(q, \omega)$ [51, 63]

$$\epsilon_{ij}(q, \omega) = \delta_{ij} - v_{ij}(q)\chi_j(q, \omega) = 0, \quad (1)$$

where $i, j = 1$ or 2 denote the two graphene layers. From Eq. (1), the two-component determinant equation becomes

$$\epsilon(q, \omega) = [1 - v_{11}(q)\chi_1(q, \omega)][1 - v_{22}(q)\chi_2(q, \omega)] - v_{12}(q)v_{21}(q)\chi_1(q, \omega)\chi_2(q, \omega), \quad (2)$$

where χ_1 (χ_2) is the non-interacting density-density response function of the top (bottom) graphene layer. Since the dielectric environment consists of three contacting media with different dielectric constants ϵ_1 ($z < 0$), ϵ_2 ($0 < z < d$) and ϵ_3 ($z > d$), the Coulomb 2D Fourier transform of the electron-electron interaction can be defined as [63]

$$v_{ij}(q, d) = \frac{2\pi e^2}{q\epsilon_{ij}(qd)} . \quad (3)$$

Here, ϵ_{ij} accounts for the three effective dielectric functions defined as

$$\frac{1}{\epsilon_{11}(q)} = \frac{2(\epsilon_2 \cosh qd + \epsilon_3 \sinh qd)}{\epsilon_2(\epsilon_1 + \epsilon_3) \cosh dq + (\epsilon_1 \epsilon_3 + \epsilon_2^2) \sinh qd} , \quad (4a)$$

$$\frac{1}{\epsilon_{22}(q)} = \frac{2(\epsilon_2 \cosh qd + \epsilon_1 \sinh qd)}{\epsilon_2(\epsilon_1 + \epsilon_3) \cosh dq + (\epsilon_1 \epsilon_3 + \epsilon_2^2) \sinh qd} , \quad (4b)$$

$$\frac{1}{\epsilon_{12}(q)} = \frac{2\epsilon_2}{\epsilon_2(\epsilon_1 + \epsilon_3) \cosh dq + (\epsilon_1 \epsilon_3 + \epsilon_2^2) \sinh qd} . \quad (4c)$$

From Eq. (3), the intra- and interlayer Coulomb interaction matrix elements are given by v_{ii} and v_{ij} , respectively, where $i, j = 1, 2$ are again the graphene monolayer indices. In order to facilitate the understanding of the plasmonic dispersion in the DLG system, we assume that the optical and acoustic plasmon dispersion attain, respectively, their long-wavelength forms defined as [63]

$$\omega_{OM}(q) = \sqrt{\frac{ge^2 v_F k_F}{\epsilon_{13}}} q , \quad (5a)$$

and

$$\omega_{AM}(q) = \frac{1 + q_{TF}d}{\sqrt{1 + 2q_{TF}d}} v_F q , \quad (5b)$$

where $\epsilon_{13} = (\epsilon_1 + \epsilon_3)/2$, $g = 4$ represents the spin and valley degeneracy in graphene, $q_{TF} = ge^2 k_F^2 / (\epsilon_2 E_F)$ is the Thomas-Fermi screening wave vector in graphene, and k_F ($E_F = v_F k_F$) is the Fermi wave vector (Fermi energy). Thus, from Eqs. (5a) and (5b), the well-known characteristic square-root dispersion ($\omega_{OM}(q) \propto \sqrt{q}$) of the optical plasmon mode and the linear dependence ($\omega_{AM}(q) \propto q$) of the acoustic plasmon mode can be observed. As discussed in Ref. 57, the concept of plasmon-phonon coupling can be understood from the basis of simple two coupled classical harmonic oscillators, where the hybrid plasmon-phonon modes are given by

$$\omega_{\pm}^2 = \frac{1}{2} \left[\omega_{ph}^2 + \omega_{pl}^2 \pm \sqrt{(\omega_{ph}^2 - \omega_{pl}^2)^2 + 4\Omega^2 \omega_{ph} \omega_{pl}} \right] . \quad (6)$$

In Eq. (6), ω_{ph} is the TMD phonon frequency, while ω_{pl} is the plasmon dispersion which can be replaced by Eq. (5a) or Eq. (5b). Ω plays the role of the coupling energy associated with the interaction between the TMD phonons and the optical or acoustic Dirac plasmons.

A. Computational method

The quantum-electrostatic heterostructure (QEH) model [64] is employed to calculate the plasmonic properties of the vdWs represented in Fig. 1(a,b). This accurate computational method is a DFT-based technique that takes into account the screening from electronic transitions at the level of the random phase approximation (RPA), where the active TMD phonon contribution of the 2D layers, screening from homogeneous bulk substrates, and the graphene doping contributions are treated with *ab initio* precision [64–66].

One of the major advantages of this method is that QEH calculates the total dielectric properties of the vdWs with layer-by-layer precision, since the contribution of each layer is considered separately. Over the last few years, QEH has been demonstrated as a very useful tool for theoretical plasmonic investigations, in various heterostructures [65–69], mostly yielding experimentally realistic results. For example, Ref. 67 presented an excellent agreement between the QEH and experimental results on heterostructures composed of graphene and hexagonal boron nitride (hBN). The authors employed the QEH method to demonstrate the possibility to probe the structure and composition of different vdWs using graphene on top of different TMDs through the sensitivity of the graphene plasmons to the dielectric environment (changing, consequently, the plasmon wavelength).

Another important advantage of the QEH model is its integration with the Computational 2D Materials Database (C2DB) [70, 71], a vast database containing the dielectric building blocks (DBBs) of 2D materials [72]. Therefore, using the C2DB results, which were previously obtained with thorough *ab-initio* calculations, allows us to detail the effects of the coupling of acoustic plasmons, that arise in double-layer graphene system [60], to active TMD phonon modes in a vdWs, without the need to treat the dielectric environment as slabs of bulk material. The fundamental assumption behind the QEH model is the additivity of independent polarizabilities of individual layers. Therefore, in plasmon-phonon coupling, the QEH model does not encompass the effects of phonon renormalization and shift. However, this approximation, remarkably accurate when there is no hybridization between neighboring layers' wave functions, remains valid even in the presence of interlayer hybridization [65]. Furthermore, the QEH model is in excellent agreement with s-SNOM experimental results [67].

Finally, to identify the acoustic plasmon-phonon coupling strength Ω , we analyze the loss function, defined as $L = -\text{Im}\{1/\epsilon(q, \omega)\}$, obtained from realistic calculations using the QEH model. In this case, due to the hybridization between acoustic plasmon and the TMD phonon, an anticrossing between upper ($\omega_+(q)$) and bottom ($\omega_-(q)$) hybrid modes emerges, close to the phonon frequency. Therefore, the coupling strength can be calculated from

TABLE I. Optical phonon frequencies for the free-standing monolayer of MoS₂ and WS₂ considered in the QEH calculations. The vibrational optical phonon modes of their monolayers are represented by E'' (R), E' (IR and R), A'₁ (R) and A'₂ (IR), where IR (R) means that the mode is active for infrared (Raman) excitations [45, 82–86].

	Phonon frequencies (meV)			
	1 (E'')	2 (E')	3 (A' ₁)	4 (A' ₂)
MoS ₂	34.19	46.35	47.59	56.16
WS ₂	35.56	42.85	50.12	51.00

the minimum energy splitting, as [57]

$$\Omega = \frac{1}{2}[\omega_+(q) - \omega_-(q)]_{\min}, \quad (7)$$

where $\omega_+(q)$ and $\omega_-(q)$ are the frequencies of the top and bottom hybrid plasmon-phonon modes. For the sake of completeness, we summarize here the main aspects behind QEH calculations based on Refs. [64] and [65]. For more details and a complete description, please see the respective references and their Supplemental Materials.

To calculate the loss function and plasmon dispersion of the vdWhs, the QEH numerically couples the density-density response function of each the i -th layer $\chi_i(z, z', \mathbf{q}_{\parallel}, \omega)$, present in the DBBs of the C2DB. To obtain the total function of the entire heterostructure, the QEH method couples each single layer together, i.e. the DBBs, through the long-range Coulomb interaction by solving a Dyson-like equation. Therefore, the Dyson equation of the total density-density response function of the complete vdWh is given by [64] $\chi_{i\alpha, j\beta} = \chi_{i\alpha} \delta_{i\alpha, j\beta} + \chi_{i\alpha} \sum_{k \neq i, \gamma} V_{i\alpha, k\gamma} \chi_{k\gamma, j\beta}$, where, for simplicity, the $\mathbf{q}_{\parallel}$ and ω variables were omitted. $\alpha = 0, 1$ represents the monopole and dipole components, respectively, and the Coulomb matrices are defined as $V_{i\alpha, k\gamma}(\mathbf{q}_{\parallel}) = \int \rho_{i\alpha}(z, \mathbf{q}_{\parallel}) \Phi_{k\gamma}(z, \mathbf{q}_{\parallel}) dz$, where $\Phi_{k\gamma}(z, \mathbf{q}_{\parallel})$ is the potential created by the density profile, $\rho_{k\gamma}(z, \mathbf{q}_{\parallel})$. Using this formalism, the inverse tensorial dielectric function of the vdWh reads

$$\epsilon_{i\alpha, j\beta}^{-1}(\mathbf{q}_{\parallel}, \omega) = \delta_{i\alpha, j\beta} + \sum_{k\gamma} V_{i\alpha, j\beta}(\mathbf{q}_{\parallel}) \chi_{k\gamma, j\beta}(\mathbf{q}_{\parallel}, \omega). \quad (8)$$

Consequently, the loss function can be found through

$$L(\mathbf{q}_{\parallel}, \omega) = -\text{Im} [\text{Tr} (\epsilon^{-1}(\mathbf{q}_{\parallel}, \omega))] . \quad (9)$$

Finally, from Eq. (9), collective modes, such as the plasmon dispersion, are obtained as the maxima of the loss function.

One important effect in vdWhs occurs when TMDs phonons couple to surface plasmon-polaritons (SPP). In this case, nontrivial effects in the (q, ω) -dispersion are originated, giving rise to one or more hybrid regions, i.e. mixed region(s) with plasmon and phonon dispersion mixed, resulting in a surface plasmon-phonon-polaritons (SPPPs). To compute this important phenomenon, i.e.

the phonon contribution of each 2D material, it is necessary to include in the computational model [65] the Γ -point phonon modes of the individual layers and the Born effective charges. The first contribution is presented in Tab. I, while the Born effective charges are defined as tensors that give the proportionality between the variation of the 2D polarization density P_i , due to an atomic displacement, defined as $Z_{i,aj} = \frac{A_{\text{cell}}}{e} \left. \frac{\partial P_i}{\partial u_{aj}} \right|_{E=0}$. Here, the A_{cell} is the in-plane area of the 2D layer, a denotes an atom, and $i-j$ are cartesian coordinates [65]. Subsequently, the lattice polarizability of a 2D material in the optical limit ($\mathbf{q} = 0$) can be obtained through [65]:

$$\alpha_{ij}^{\text{lat}}(\omega) = \frac{e^2}{A_{\text{cell}}} \sum_{ak, bl} Z_{i,ak} [(\mathbf{C} - \mathbf{M}(\omega^2 - i\gamma\omega))^{-1}]_{ak, bl} Z_{j,bl}, \quad (10)$$

where $\mathbf{C}$ is the force constant matrix in the optical limit, $\mathbf{M}$ is a diagonal matrix containing the atomic masses, the broadening parameter γ is set to 10 meV [71], representing the phonon lifetime (see the *Supporting Information* of Ref. [65] for a derivation of Eq. 10).

Thus, considering the contributions of electrons and phonons, the total monopole and dipole component of the DBB of layer i are defined as [65]

$$\chi_{i0}^{\text{total}}(\mathbf{q}_{\parallel}, \omega) = \chi_{i0}^{\text{el}}(\mathbf{q}_{\parallel}, \omega) - \mathbf{q}_{\parallel}^2 \alpha_{\parallel\parallel}^{\text{lat}}(\omega) \quad (11a)$$

$$\chi_{i1}^{\text{total}}(\mathbf{q}_{\parallel}, \omega) = \chi_{i1}^{\text{el}}(\mathbf{q}_{\parallel}, \omega) - \alpha_{zz}^{\text{lat}}(\omega), \quad (11b)$$

where $\alpha_{\parallel\parallel}^{\text{lat}}$ denotes the 2×2 in-plane submatrix of α^{lat} . All DBBs in this work are from the C2DB project. The electronic polarizability χ_{i0}^{el} calculation is based on random phase approximation (RPA) [70], while lattice polarizability α_{ij}^{lat} is calculated in the recent progress of the C2DB [71] and the Ref. [65].

B. The coupling strength regime

In order to investigate how significant the plasmon-phonon hybridization is, it is necessary to compare the plasmon-phonon coupling strength (Ω) to other relevant energy scales, for example, the phonon energy ($\hbar\omega_{ph}$) and the linewidth of the coupled system [73]. In this situation, the splitting becomes significant only when the coupling Ω exceeds the linewidths of the coupled systems, which also enables the experimental observation of these two modes. Thus, if Ω is very small compared to other important energy scales, for example, the phonon energy, the coupling is negligible and is not strong enough to change the original (uncoupled) frequencies. This defines different coupling regimes. The first one, where Ω can effectively be neglected, is classified as weak coupling (WC) [73, 74]. On the other hand, if the strength of the plasmon-phonon coupling is large enough to modify the uncoupled energy dispersion, thus creating hybrid

plasmon-phonon modes, the coupling regime can be classified as strong (SC) [73, 74]. The stronger coupling reflects more efficient plasmon-phonon interactions, which is crucial for many applications based on, or mediated by, hybrid states [75–77]. In this article, we define the coupling regimes in a pragmatic way, after normalizing the coupling strength to the phonon frequency at the source of hybridization, as $\eta = \Omega/\hbar\omega_{ph}$. Then, the WC regime corresponds to $\eta < 0.01$, and SC regime to $\eta \geq 0.01$. In addition, and as discussed in Ref. 75, we define the ultrastrong coupling (USC) regime for $\eta \geq 0.1$ - where plasmon-phonon coupling not only alters the characteristic energy dispersion, but may radically change the observable properties of the system [78].

C. Electric field profile in double-layer graphene intercalated with a TMD

Interaction between confined modes localized at each of the two interfaces in the DLG system forms coupled modes. To show the electric field profile of such coupled modes in TMD-intercalated double-layer graphene, the finite-element method (FEM) is used to perform full-wave simulations. In these simulations, the TMD-intercalated double-layer graphene is placed in the center of the computational domain, surrounded by a perfectly

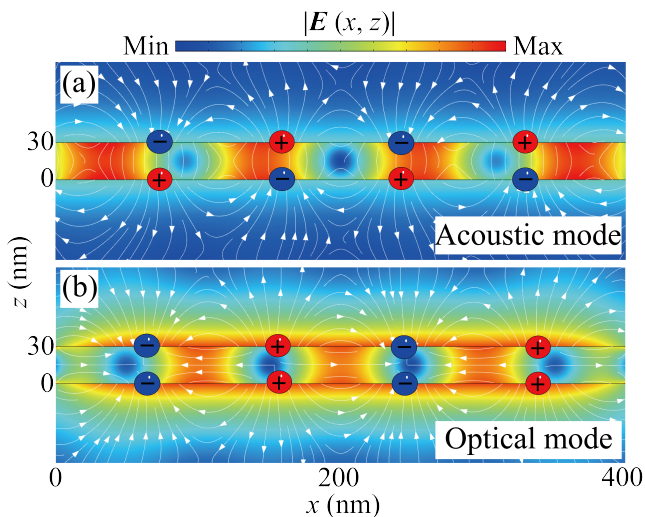


FIG. 2. (Color online) Schematic illustration of the electric field lines $\mathbf{E}(x, z)$ in a double-layer graphene system, with the color map showing the normalized magnitude of the field ($|\mathbf{E}(x, z)|$). The top and bottom layers are separated by $d = 30$ nm. For simplicity, the dielectric constant of the top and bottom layer is $\epsilon_1 = \epsilon_2 = 1$, and in-between $\epsilon_3 = 4$. Panel (a) stands for the acoustic mode (AM) and (b) for the optical mode (OM). The positive (red circles) and negative (blue circles) charges represent, qualitatively, the formation of dipole like-charges. For the AM, the electric fields in-between the layers (red regions) increase the coupling of plasmons to the out-of-plane optical phonon modes.

matched layer. Graphene can be considered as a conducting interface [87], or a boundary condition that sustains the confined electronic-magnetic field mode, with a simple Drude surface conductivity as [88]

$$\sigma(\omega) = \frac{e^2 E_F}{\pi \hbar^2} \frac{i}{\omega + i\tau^{-1}}. \quad (12)$$

In Eq. (12), we consider $E_F = 100$ meV as the Fermi level of the graphene and $\tau \sim 10^{-13}$ s as the relaxation time for electrons. Under a normal-incidence plane-wave $\mathbf{E} = E_0 e^{i(k_0 z - \omega t)} \hat{x}$, x- and z- component of electric fields are plotted in Fig. 2, considering a TMD thickness $d = 30$ nm, representing the distance between two graphene layers.

III. RESULTS AND DISCUSSION

A. Overview of plasmon-phonon dispersion in double layer graphene-based vdWhs

Fig. 3(a) shows the loss function obtained from the QEh for a G/10-MoS₂/G heterostructure. In this figure one observes that the plasmon dispersion presents both optical ($\hbar\omega_{OM} \propto \sqrt{q}$) and acoustic ($\hbar\omega_{AM} \propto q$) surface plasmon-phonon polaritons (i.e. SPPPs), and four MoS₂ phonon modes (the last mode is magnified in the inset of panel (b)). It is also possible to observe, from the loss function, a well-defined split mode (anticrossing) for the coupling between acoustic plasmon mode (AM) and A_2'' phonon mode, in contrast to the optical mode (OM). This means that the coupling is much stronger for the acoustic than for the optical case. Since our interest is in the acoustic plasmon-phonon coupling, Fig. 3(b) emphasizes the plasmon dispersion close to the hybridization region. Fig. 3(c) shows a magnification of the square box marked in panel (b). From this panel, two clearly split hybrid modes are seen, labeled as ω_+ and ω_- , where the point of minimum energy difference between them gives us the coupling strength Ω , as presented in Fig. 3(d). The dashed white line in Figs. 3(b,c), used for reference, represents the surface plasmon polariton (SP²) dispersion without considering the phonon contribution, i.e taking into account only the dielectric contributions of the TMD.

Under the same excitation momentum, the frequency of the acoustic mode is lower, that is, closer to the phonon frequencies. In addition, the electric field of the acoustic plasmon mode becomes more localized between the two graphene layers, which is also demonstrated in the simulation shown in Sec. II C. It is important to point out that, although the TMD layers have four non-degenerate optical phonon modes, only A_2'' (IR) are out-of-plane IR-active phonon modes, as shown in Fig. 1(c). Due to the out-of-plane phonon vibrations and dipolar-like acoustic plasmon electric field mode, A_2'' (IR) phonons match the acoustic plasmon mode perfectly to enhance the plasmon-phonon coupling between them.

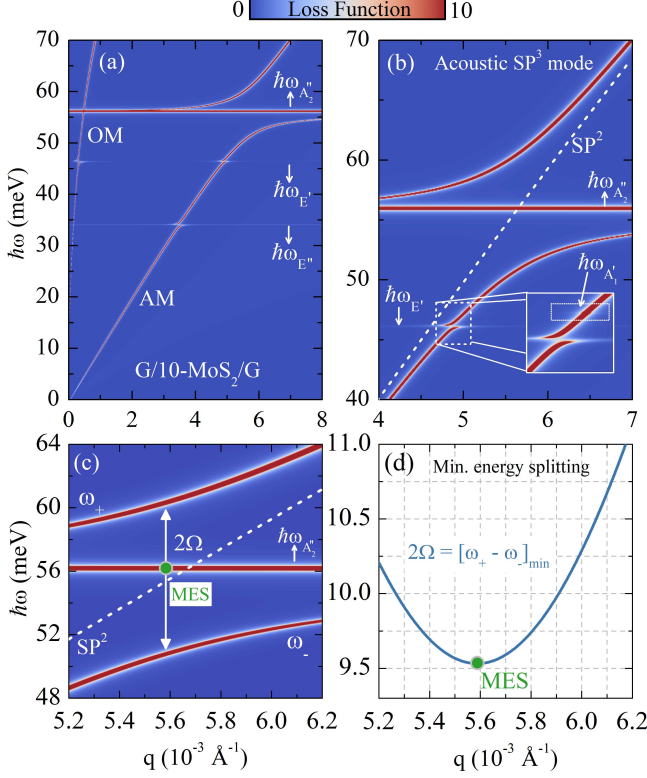


FIG. 3. (Color online) (a) Loss function of the surface plasmon-phonon polariton dispersion for a G/10-MoS₂/G vd-Whs with $E_F = 150$ meV. OM and AM indicate the optical and acoustic Dirac plasmon modes, respectively. Horizontal branches in the loss function are signatures of the four MoS₂ phonon modes, labeled as E'' (R), E' (IR and R), A₁' (R) (see magnification in panel (b)) and A₂' (IR). IR (R) means that the mode is active for infrared (Raman) excitations. (b) The same as in (a), but now emphasizing the coupling between the acoustic Dirac plasmons mode (AM) and the MoS₂ phonon modes. (c) Magnification close to the IR-active A₂' phonon mode where two well-defined hybrid modes ω_+ and ω_- arise due to the plasmon-phonon coupling. (d) Minimum energy splitting (MES), defined as $2\Omega = [\omega_+ - \omega_-]_{\min}$, extracted from the results in panel (c). Ω represents the plasmon-phonon coupling strength. The white dashed lines in panels (b) and (c) show the plasmon dispersion without the contribution of TMD phonons (SP²), for reference.

B. The influence of the number of TMD layers on the plasmon-phonon coupling strength

Fig. 4(a) shows the acoustic plasmon-phonon coupling strength Ω as a function of the number of TMD layers (N) in-between the graphene monolayers, for $E_F = 100$ meV. Blue lines and symbols stand for G/ N -MoS₂/G results, while orange lines and symbols are for G/ N -WS₂/G. At first, the acoustic SPPs coupling (Ω) increases with the number of MX₂ layers, but after reaching a maximum value (i.e. near $N = 25$ layers), it starts to slowly decrease, and finally converges to a value, which is the bulk limit of this coupling parameter. The ob-

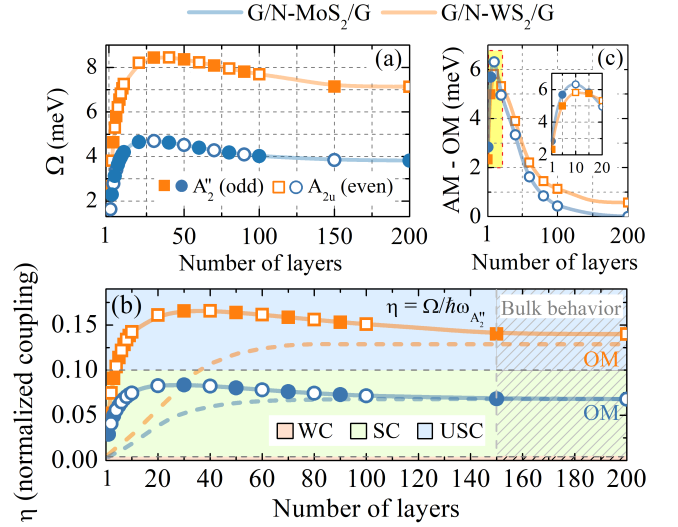


FIG. 4. (Color online) Acoustic surface plasmon-phonon polaritons (ASPPPs) coupling strength (Ω) as a function of the number of TMD layers (N) in-between two graphene monolayers (cf. Fig. 1(a, b)), for $E_F = 100$ meV. Blue lines and dots (orange lines and squares) refer to N -MoS₂ (N -WS₂) results. (a) Coupling energy between acoustic Dirac plasmon and the IR-active out-of-plane A₂' (solid symbols) and A_{2u} (open symbols) phonon modes, for odd and even number of layers, respectively. (b) ASPPPs coupling strength normalized to the respective monolayer phonon frequencies at Γ -point defined as $\eta = \Omega_{1(2)}/\hbar\omega_{A_2'}$. Three different regions, blue, green, and pink, represent the WC ($\eta < 0.01$), SC ($0.01 \leq \eta < 0.1$), and USC ($\eta \geq 0.1$) regime, respectively [75]. Blue and orange dashed lines are added for reference and represent the optical mode of a G/ N -MX₂ vdWhs with $E_F = 100$ meV [57]. The shaded area represents the bulk limit of the ASPPPs coupling, reached for approximately 150 TMD layers. In this case, the acoustic plasmon mode converges approximately to the optical mode computed in Ref. 57, i.e. for a mono-graphene-based vdWhs. (c) Difference between the acoustic and optical normalized plasmon-phonon coupling in DLG-based vdWhs and mono-graphene-based vdWhs, respectively.

served non-monotonic trend is due to the interplay between the available phonon modes and the confined electric field induced by plasmon-plasmon interaction in the two graphene layers, which are two competing effects. With the increase of the number of MX₂ layers, more oscillators are involved, i.e. more phonons are available to couple with Dirac plasmons [79, 80]. On the other hand, as the number of layers is increased further, the distance between the two graphene sheets increases, which leads to a weaker confined electric field between them. The two different coupling mechanisms, which depend on phonon modes involved and the confined electric fields, will dominate in different ranges of the number of TMD layers and thus determine the relationship between coupling strength and the bulk limit.

C. SPPP interaction: weak, strong and ultra-strong coupling regime, *vs.* the bulk limit

Fig. 4(b) shows the normalized acoustic SPPPs coupling η as a function of N -MoS₂ and N -WS₂ layers, where $\eta = \Omega/\hbar\omega_{A_2''}$, as defined by Kockum *et al.* [75], quantifies the coupling strength. Here $\omega_{A_2''}$ is the out-of-plane A_2'' phonon frequency of a single TMD layer. The results calculated in Ref. 57 for the coupling between optical plasmons (OM) arising from single-layer graphene and TMD phonons are shown in Fig. 4(b) as dashed blue (G/ N -MoS₂) and orange (G/ N -WS₂) lines, for comparison. For reference, three different regions, pink, green, and blue, are illustrated in Fig. 4(b) and represent the WC ($\eta < 0.01$), SC ($0.01 < \eta < 0.1$) and USC ($\eta > 0.1$) regimes, respectively [75]. The acoustic SPPPs coupling is stronger than the optical SPPPs coupling in both MoS₂ and WS₂ vdWhs systems.

A remarkable result for this type of vdWhs, which is evident in Fig. 4(b), is the enhancement of the coupling between acoustic Dirac plasmons and the IR-active out-of-plane A_2'' , especially for few TMD layers, in contrast to the OM in G/TMD system [57]. We consider the bulk limit is reached for $N > 150$, as all results remain unchanged for larger N . In fact, the difference between the acoustic and optical plasmon-phonon coupling, defined as $\Omega(\text{AM}) - \Omega(\text{OM})$, which is illustrated in Fig. 4(c), exhibits a maximum around ten TMDs layers (cf. the inset). Although the difference in the bulk limit for the MoS₂ case is not zero, which is related to the choice of the Fermi energy made for these calculations, this value is below 1 meV for 200 WS₂ layers. The effect of the Fermi energy on the plasmon-phonon coupling will be discussed in more detail later in this paper.

D. Acoustic *vs.* optical plasmon-phonon coupling in the bulk limit

Fig. 5 illustrates the evolution of the acoustic and optical plasmon dispersions obtained from the QEH as the number of TMD layers is increased, in order to understand the bulk limit of the G/ N -TMDs/G vdWhs. As follows from Figs. 5(a, b), calculated in a region close to the A_2'' phonon frequency, increasing the number of TMD layers converges the OM and AM modes to the same curve, which corresponds to the plasmon mode in the bulk limit. In other words, while in Fig. 5(a), for G/50-MoS₂/G, the loss function peaks for upper and bottom OM and AM are distinguishable, in Fig. 5(b), for G/150-MoS₂/G, they form approximately degenerate SPPP modes. Fig. 5(c) presents the plasmons dispersion curves for G/ N -MoS₂/G, with $N = 10$ (green), 50 (orange), and 150 (blue) in the absence of phonon contributions, i.e. only taking into account the MoS₂ dielectric constant corresponding to each of the considered thicknesses. For reference, we calculated the plasmon dispersion for graphene on top of bulk MoS₂ using the

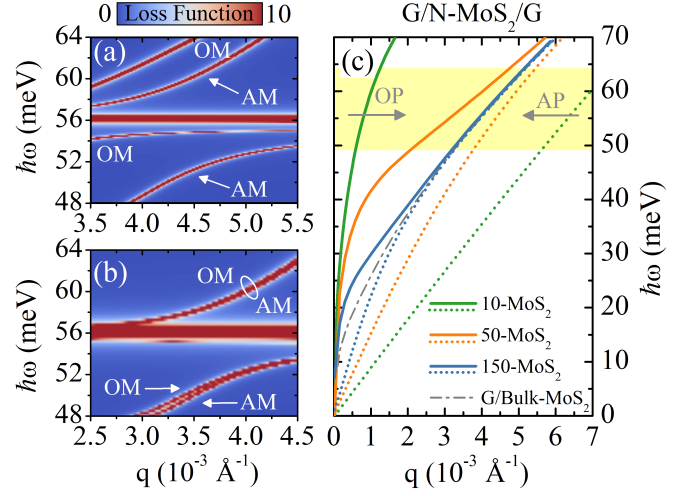


FIG. 5. (Color online) (a) Loss function of SPPPs for G/50-MoS₂/G and (b) for G/150-MoS₂/G, for $E_F = 100$ meV. OM (AM) indicates the optical (acoustic) hybrid plasmon-phonon branch. (c) Acoustic SPPs dispersion for G/ N -MoS₂/G in this case, considering just the dielectric contribution of the MoS₂ layers, i.e. disregarding the phonon contribution. Solid lines represent the optical plasmon dispersion and dotted lines stand for the acoustic plasmon dispersion. As the number of MoS₂ layers is increased, both the optical and acoustic plasmon dispersions converge to the bulk dispersion (gray dashed-dotted line). The bulk dispersion was obtained for graphene on bulk-MoS₂, with $E_F = 100$ meV. The yellow region in panel (c) corresponds to the frequency range of panels (a)-(b), emphasizing that for 150 MoS₂ layers the OM and AM become completely indistinguishable.

QEH model, represented by the gray dashed-dotted line in Fig. 5(c). One easily verifies that as the number of layers is increased, OM (solid lines) and AM (dotted lines) converge to each other and to the bulk limit (dashed-dotted line), as expected.

E. Effect of doping on the surface acoustic plasmons-polaritons

Effects of gating or doping in vdWhs can be considered in a first approximation through a corresponding shift of the Fermi energy. Figs. 6(a, b) shows how the graphene Fermi energy affects the coupling strength of G/20-MoS₂/G and G/20-WS₂/G systems, respectively. The normalized coupling strength (η) is calculated as a function of the Fermi energy (E_F), which is considered the same in both top and bottom graphene layers. As seen in the figure, in both cases the coupling strength quickly increases to the USC regime with the increase of the Fermi energy, converging to a maximum value, represented by the dotted line. As explained in Ref. 57, for low Fermi energy, the Pauli blocking is lifted and inter-band single-particle processes are allowed. As a consequence, the plasmon lifetime is inhibited.

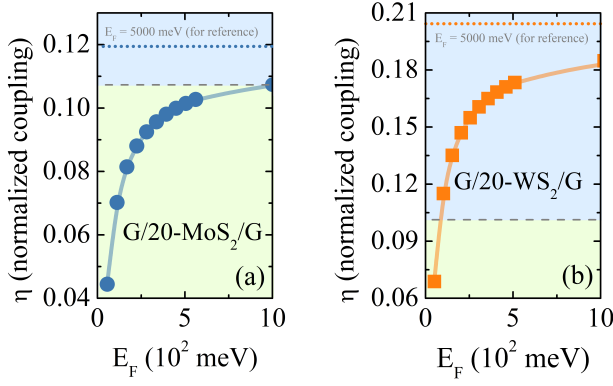


FIG. 6. (Color online) Tuning the acoustic plasmon-phonon coupling strength Ω by varying the Fermi energy simultaneously in the top ($E_F^T = E_F$) and the bottom ($E_F^B = E_F$) graphene layers. The acoustic SPPP coupling Ω is shown as a function of the Fermi energy for (a) G/20-MoS₂/G and (b) G/20-WS₂/G. The blue (orange) dotted lines in (a) [(b)] are the reference of couplings for $E_F = 5000$ meV. The green and blue regions represent SC ($0.01 \leq \eta < 0.1$) and USC ($\eta \geq 0.1$) coupling regimes, respectively.

ited, thus suppressing the plasmon-phonon coupling. In addition, the plasmon frequencies increase with the doping of graphene. The dependence of the acoustic plasmon frequency $\omega_{AM}(q)$ in DLG system on the carrier density (n_e) is $\omega_{AM}(q) \sim (1/2)n_e^{1/4}$ [51], half of the one for single-layer graphene [81]. Since the Fermi energy in graphene is related to the carrier density through the relation $n_e = (E_F/\hbar v_f)^2/\pi$, at high doping levels, the change of the acoustic plasmon dispersion in the (q, ω) -plane with the Fermi energy is not significant, which causes the flattening of the curve for high Fermi energy. Recent research has revealed that chemical doping introduces additional damping channels that can impact the plasmonic behavior of graphene [89]. Furthermore, doping also induces alterations in the phonon behavior of graphene [90, 91], thus the plasmonic response is modified via the plasmon-phonon coupling in graphene [92, 93]. To mitigate this effect, it is crucial to ensure that the intrinsic graphene plasmon energy lower than the optical phonons of graphene, which are around 0.2 eV [9, 10, 89]. Nevertheless, these results clearly show that the coupling strength can be conveniently tuned by doping.

Finally, we explore asymmetric doping, i.e. considering different Fermi energies for the top (E_F^T) and bottom (E_F^B) graphene layers (see inset in Fig. 7), to provide precise control of the coupling strength. To do so, we calculate the normalized coupling strength η for a G/20-WS₂/G vdWhs, considering a fixed $E_F^T = 100$ meV and varied values of E_F^B . Results are shown in Fig. 7. We observe that $E_F^B < 30$ meV does not generate acoustic SPPs due to Landau damping, therefore, results will be discussed only for $E_F^B > 30$ meV. As expected, when E_F^B is low, the normalized coupling strength has almost the same value as that for a G/20-WS₂ mono-graphene

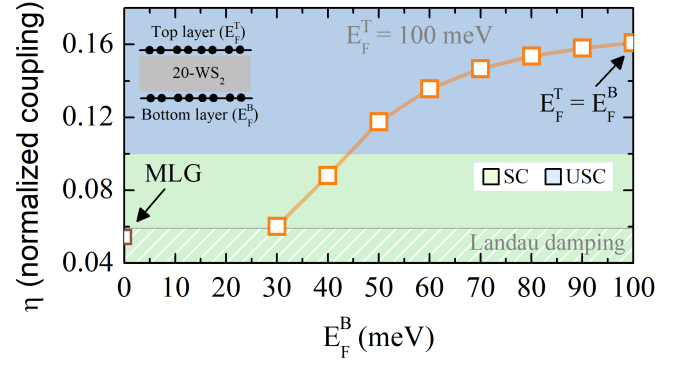


FIG. 7. (Color online) Normalized coupling strength of acoustic SPPs in double-layer graphene intercalated by 20 layers of WS₂ (the inset shows a sketch of this system), as a function of the Fermi level of the bottom graphene layer E_F^B . For convenience, the top graphene Fermi energy is fixed at $E_F^T = 100$ meV. For low E_F^B , the system behaves effectively as monolayer graphene over 20 WS₂ layers, where the acoustic SPPP is in the SC regime. The green and blue regions represent SC ($0.01 \leq \eta < 0.1$) and USC ($\eta \geq 0.1$) coupling regimes, respectively.

system (shown under the label MLG in Fig. 7, for comparison), which is in the SC regime. However, as E_F^B increases, the normalized coupling strength crosses from the SC to the USC regime at $E_F^B \approx 45$ meV, which gives a guideline to precisely fine tune the coupling strength by doping, which can be achieved even electronically, by gate voltage [31].

IV. CONCLUSIONS

We have demonstrated the possibility of tuning the coupling strength of the acoustic graphene plasmons in a double-layer graphene (DLG) with the IR-active out-of-plane A_2'' phonon modes of the N intercalated layers of MoS₂ or WS₂, thereby enabling precise control over the coupling regime ranging from weak all the way to ultra-strong coupling. The strong interaction between the plasmonic and phononic modes in such G/ N -MX₂/G heterostructures leads to the formation of hybrid excitations with unique optical and electronic properties. Our findings show that, for a limited number of TMD layers, the coupling strength between acoustic plasmons and phonons in DLG-based van der Waals heterostructures surpasses that of optical plasmons in mono-graphene-based heterostructures discussed earlier. Our numerical simulations reveal a peak at $N = 10$ layers that differs 6 meV between single and double-layer cases. This is due to the dipole-like nature of acoustic graphene plasmons, as explained in Sec. II C.

Our calculations also show that, for $E_F = 100$ meV, the coupling energy converges to a constant (bulk limit) for a number of TMD layers larger than $N = 150$, where the acoustic and optical plasmon-phonon couplings con-

verge to the same value, which is also verified by comparing the loss functions of G/50-MoS₂/G and G/150-MoS₂/G. In the latter, one can no longer distinguish between the optical and acoustic plasmon dispersions, since the top and bottom graphene layers are far apart.

Finally, we discussed the effect of doping on the surface acoustic plasmon-polaritons, changing the Fermi energy in both the top (E_F^T) and bottom (E_F^B) graphene layers of the studied G/*N*-MX₂/G heterostructures. We have demonstrated that different values of E_F^T and E_F^B enable one to tune the coupling parameter from strong to ultra-strong coupling regimes, thus rendering such asymmetric doping a fine adjustment knob for the coupling regimes of this system. Even though we presented results only for two selected TMD materials, our findings are easily transferable for plasmon-phonon coupling phenomena in different DLG-based vdWhs. The precise yet broad tuning of coupling found here, ranging from weak to ultra-strong regimes, may be a key achievement towards novel nano-thermoelectronics and on-chip optical devices, as well as further breakthroughs in understanding the behavior

of materials at the nanoscale.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study will be made available by the authors upon reasonable request.

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