

Fast two-dimensional long-wavelength plasmons in atomically thin MoS₂

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Plasmons are bosonic wavelike charge oscillations in solids and the plasmon group velocity (v_g), the first derivative of momentum (q)-dependent dispersions, depicts the many-body electron motions of critical electronic importance. The profound characteristics of v_g in either three-dimensional (3D) or two-dimensional (2D) electron systems are nonetheless short of quantitative scrutiny. Using q -resolved electron energy loss spectroscopy, we tackle the q -dependent v_g of π and $\pi + \sigma$ plasmons in 3D-bulk and 2D-monolayer (ML) MoS₂ semiconductors, with the 2D MoS₂ being the model transition-metal dichalcogenide (TMD) for emergent atomically thin electronics. v_g along with the underlining electronic parameters of the charge density, effective mass, and screening length in 2D for the plasmon dispersions is quantitatively elaborated. 2D v_g is further compared to those evaluated from metallic ML-TMD TaS₂ and graphene, and the scaling over the single-particle Fermi velocity (v_F) unveils a v_g/v_F function generally obeyed by the semiconducting and metallic TMD and graphene of distinctly different massive and relativistic characters, respectively. Beyond the pertinent critical q (on the order of $10^{-2} \text{ \AA}^{-1}$) where one observes $v_g = v_F$, electric-field lines connecting the oscillating 2D charges would turn from vacuum into the atomically thin matters, concomitant with the increasing plasmon damping and reducing v_g ($v_g < v_F$). Long-wavelength 2D plasmons below the critical q characterize the field lines largely in vacuum and are persistent and fast ($v_g > v_F$), with high-speed application merits. Conversely, 3D plasmons in bulk MoS₂ never exceed v_F .

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I. INTRODUCTION

Pointed out by D. Pines and D. Bohm in the seminal work on collective, long-range Coulomb interactions in solids of a three-dimensional (3D) free-electron-gas (FEG) essence, dynamical motions of charges are spontaneously screened by concomitantly fluctuated charge densities as if the charges as a whole are oscillating in the absence of external electromagnetic stimuli [1]. Such collective oscillations of charges, dubbed plasmons with the quanta of $\hbar\omega_p^{3D}$ ($\hbar$ is the reduced Planck's constant and ω_p^{3D} is the plasmon frequency), represent the hallmark of many-body physics [1–3] and also a form of bosons [4,5].

In the classical 3D context of massive fermions, mobile conduction electrons and bound valence electrons equally

facilitate the bosonic oscillations, with $\omega_p^{3D} = \sqrt{\frac{4\pi n_{3D}e^2}{m^*\epsilon_\infty}}$ (n_{3D} is the 3D charge density, e is the electron charge, m^* is the effective mass, and ϵ_∞ is the screening dielectric constant) [1–3]. Upon the characteristic q^2 dispersion, the 3D-plasmon energy matches the single-particle continuum of individual charges at the critical q_c^{3D} of ω_p^{3D}/v_F (on the order of $0.1 \text{ \AA}^{-1}$) and decays into electron-hole pairs, narrated as the Landau damping [3,6]. These many-body physics receive extensive q -dependent spectroscopy investigations through decades [5,7–11], with the recent breakthrough that the 3D-plasmon dispersion devises the direct measurement of m^* and v_F of constituent fermions across a charge-density-wave transition [10].

The interest in plasmons delves into two-dimensional (2D) systems of atomically thin matters [12–16]. Within the classical 2D-FEG framework, the 2D plasmon disperses by $\sqrt{q}$ and is gapless in the long-wavelength limit as $\omega_p^{2D} = \sqrt{\frac{2\pi n_{2D}e^2}{m^*\epsilon_\infty}} \sqrt{q}$ [12–14], in contrast to the finite ω_p^{3D} upon the vanishing q [9]. The q -resolved attempts to tackling the 2D plasmons turn into realities by graphene [17–19], the relativistic nature of which is, however, at odds with the massive-electron essence of ω_p^{2D} . The reported Dirac- and π -plasmon dispersions therein in graphene [17–19] shall, therefore, be delineated by

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quantum-mechanical $\omega_p^{\text{QM}} = e\sqrt{\frac{v_F}{\varepsilon_\infty \hbar}}(g\pi n_{2D})^{\frac{1}{4}}\sqrt{q}$ (g is the degeneracy factor of $g = g_s g_v$, and g_s and g_v are the respective spin and valley degeneracies [20,21]) [14] and are yet thus scrutinized. The massive electron system of 2D-monolayer (ML) transition-metal dichalcogenide (TMD) otherwise offers the opportunity for elaborating on ω_p^{2D} . Intriguingly, the recent theoretical and experimental q -resolved studies of the 2D plasmon of mobile carriers in metallic ML TaS₂ indicate a flattened ω_p^{2D} dispersion in $\sqrt{\frac{q}{\varepsilon_\infty}}$, where ε_∞ is nonlocal due to the interband-transition screening [15,22]. The flattened dispersion leads to the suggestion of a characteristically slow v_g [15,22] and pervasive interband transitions across versatile ML TMD are proposed to cast generally slow v_g in the 2D TMD [15]. This slow v_g notion, short of q -dependent inspections so far, is highly relevant considering the broad interest in ML TMD for fast electronics [23]. Moreover, the flattened 2D plasmon was theoretically suggested to be undamped by the avoided crossing with the single-particle decay channel [15], while the experimental scrutiny revealed a damped character, for which the origin remains unresolved [22] and deserves our attention.

Here, we report on the quantitative q -resolved electron energy loss spectroscopy (q -EELS) study of the v_g of π and $\pi + \sigma$ plasmons in the freestanding TMD semiconductor of MoS₂ in 3D-bulk and 2D-ML forms free from intrinsic charge carriers [24–26]. In analogy to graphene, the valence electrons in MoS₂ are of π and $\pi + \sigma$ characters [27–29]. There have been experimental reports on the π and $\pi + \sigma$ plasmons [7,8,11,28,30–32], while the thorough dispersions from the light line ($q \rightarrow 0$) are left unexplored. We reveal the 3D- and 2D-plasmon dispersions from $q \rightarrow 0$ (q resolution $\sim 0.001 \text{ \AA}^{-1}$) to zone boundaries, establishing their deviations from the FEG ω_p^{3D} and ω_p^{2D} by ubiquitous interband transitions. In particular, the 2D plasmons are gapped at the light line, attributed to fermionic interband transitions at the respective light-line crossovers. Departing from the light line, the 2D excitations gain bosonic dispersions, manifesting the distinct fermionic-to-bosonic turnover, and obey the proposed nonlocal dispersion of $\sqrt{\frac{q}{\varepsilon_\infty}}$ with $\varepsilon_\infty = 1 + \rho q$ (ρ is the charge screening length) [15]. ρ is quantitatively derived ($\sim 12 \pm 0.6 \text{ \AA}$) and v_g is evaluated against the single-particle limit of v_F in 2D, unraveling that the persistent 2D plasmons are long wavelength with $q < q_c^{\text{2D}}$ ($q_c^{\text{2D}} = 1/\rho$ [15] and the concomitantly observed $v_g = v_F$) and fast ($v_g > v_F$). The suggested slow 2D plasmons appear at $q > q_c^{\text{2D}}$ with $v_g < v_F$ and would be effectively damped. The general scaling of $\frac{v_g}{v_F} = \frac{e}{2\hbar} \sqrt{\frac{m^* g_v}{\varepsilon_\infty q}}$ in the 2D TMD and graphene, and the 3D plasmons are discussed.

II. METHOD

The 3D-bulk MoS₂ for q -EELS was prepared by mechanical exfoliations down to the thickness of $\sim 30 \text{ nm}$ and the 2D-ML MoS₂ studied was grown on sapphire by chemical vapor deposition and then transferred onto a Cu grid in the freestanding form. The q -EELS experiments were performed on a field-emission transmission electron microscope (TEM;

FEI Tecnai F20), operated at 200 keV and equipped with an EELS spectrometer (energy resolution 0.54–0.6 eV). To accomplish the superb q resolution on the order of $\sim 0.001 \text{ \AA}^{-1}$ required for revealing plasmons close to the light line [19], we exploited the Midgley method [33], which considers the sample itself as the optical grating and diffracts the incident focused electron probe by significant elevations of the specimen from the eucentric plane by $\sim 350 \text{ \mu m}$ in maximum that is equivalent to the sample defocus length (z). The electron optics were set in the nanoprobe mode for the optimal beam coherence and minimal beam damage. The corresponding electron-beam size (d) is $\sim 4 \text{ \AA}$ and the concomitant beam-convergence semiangle (α) is $\sim 3 \text{ mrad}$, with $\alpha = \pi/dk_0$ ($k_0 = 2\pi/\lambda_0$; the electron wavelength λ_0 at 200 kV is $0.0251 \text{ \AA}$ [34]) [22]. The maximal z of $\sim 350 \text{ \mu m}$ corresponds to the radius (r) of the illuminated area (A), $\sim 1.1 \text{ \mu m}$ by $r = z\alpha$ ($A \sim 4.5 \text{ \mu m}^2$). By the nanoprobe current of $\sim 12 \text{ pA}$, the corresponding electron dose on the ML MoS₂ is $\sim 1.6 \times 10^7 \text{ e}/\text{\mu m}^2$, much smaller than the ubiquitous electron dose on the order of $10^7 \text{ e}/\text{\AA}^2$ in EELS in the scanning TEM (STEM) mode [29] and advantageous for reducing beam-induced carbon contaminations and damages.

For accessing the light-line regime, we multiply z by optical magnifications [19], reaching the diffraction projection length of $\sim 26 \text{ m}$, and the full width at half maximum of the corresponding diffraction spots is as sharp as $\sim 0.0009 \text{ \AA}^{-1}$, which corresponds to the associated q resolution [35]. Using the q -selection slit of $2 \text{ mm} \times 0.3 \text{ mm}$, we then acquired the q -EELS spectral intensity along the 0.3-mm slit-width direction and the q dispersion was registered in the 2-mm slit-length direction (binning 2 for optimal spectral intensities) using a TEM sample-rotation holder to orient the ΓK (ΓM) along the slit length, leading to the achievement of an ω - q map over a given q range [e.g., the light-line regime of Figs. 1(a) and 1(b); the pixel size in q is $\sim 0.00024 \text{ \AA}^{-1}$] [35]. To ease the collection of weak q -EELS spectra beyond $\sim 0.02 \text{ \AA}^{-1}$, three shorter projection lengths were separately adopted and increased the spectral intensity by coarsening the pixel size [35]. The individual q -EELS spectra shown below $\sim 0.02 \text{ \AA}^{-1}$ are the integration from Figs. 1(a) and 1(b) over five pixels and those above $\sim 0.02 \text{ \AA}^{-1}$ are the five- and three-pixel integrations over the corresponding ω - q maps [35]. Integration over the odd-number pixels has been based on the consideration of the use of the middle pixel for denoting the designated q and, meanwhile, the integrated q scale is always comparable to the related q resolution to warrant the preservation of the q resolution. For instance, the integration of five pixels in Figs. 1(a) and 1(b) results in the integrated q of $\sim 0.0012 \text{ \AA}^{-1}$, which is close to the pertinent resolution of $\sim 0.0009 \text{ \AA}^{-1}$ [35].

First-principles calculations were performed using Quantum ESPRESSO within density-functional theory (DFT) [36] with the generalized gradient approximation (GGA) for the exchange-correlation functional [37]. No spin-orbit coupling (SOC) was included in the calculations. For monolayer MoS₂, a $12 \times 12 \times 1$ Monkhorst-Pack k -point grid was used with an in-plane lattice constant of $3.16 \text{ \AA}$ [38] and a vacuum spacing of more than $15 \text{ \AA}$ to avoid spurious interlayer interactions in a supercell configuration. Bulk MoS₂ calculations employed a $12 \times 12 \times 3k$ -point grid with experimental lattice constants $a = b = 3.16 \text{ \AA}$ and $c = 12.29 \text{ \AA}$ [38]. Both monolayer and

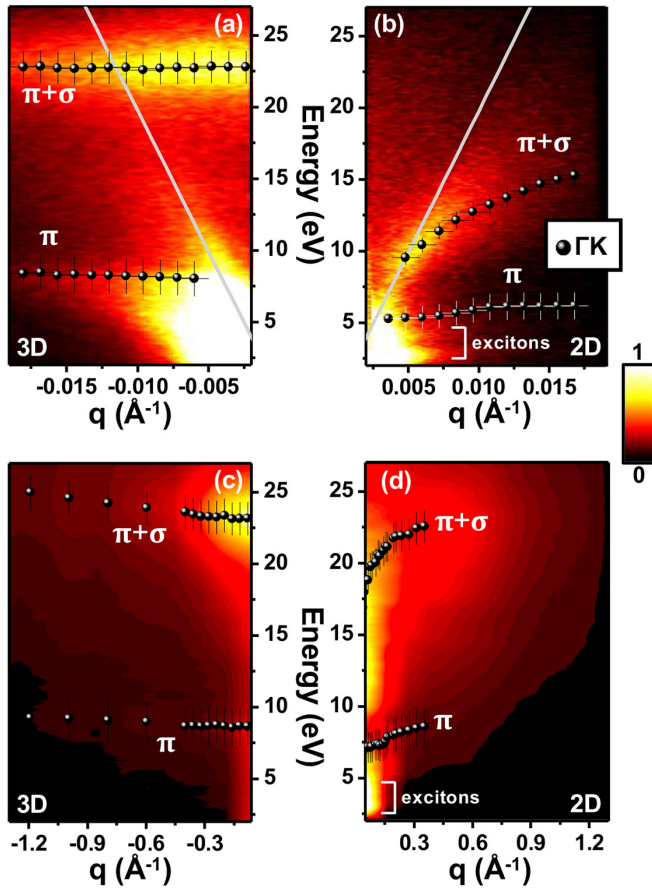


FIG. 1. (a), (b) the respective q -EELS maps of π - and $\pi + \sigma$ -plasmon dispersions along ΓK in 3D and 2D MoS₂ close to the light line (gray; map, $\sim 0.00024 \text{ \AA}^{-1}/\text{pixel}$). (c), (d) the respective 3D- and 2D-plasmon dispersions up to the ΓK zone boundary. Color scale bar: the normalized spectral intensity. The surface π and $\pi + \sigma$ plasmons in 3D are diminishingly weak and omitted for simplicity. Black dots: the pseudo-Voigt fitted plasmon peaks of the spectra integrated over designated numbers of pixels [35]. Error bars in energy: $\sim 5\%$ for the 3D π and $\pi + \sigma$ plasmons and $\sim 15\%$ ($\sim 5\%$) for the 2D π ($\pi + \sigma$) plasmon. Error bars in q : $\sim 0.001 \text{ \AA}^{-1}$ in panels (a) and (b), and ~ 0.006 and $\sim 0.02 \text{ \AA}^{-1}$ in panels (c) and (d) (see Ref. [35] for details).

bulk calculations used a plane-wave cutoff energy of 90 Ry. The complex dielectric function and EELS spectra were computed within the random-phase approximation (RPA) using the BerkeleyGW package [39], with a plane-wave cutoff of 15 Ry and inclusion of 120 electronic bands; Gaussian broadening of 1.5 eV accounted for lifetime effects. Although GW self-energy corrections would improve the bandgap and high-energy optical transition accuracies, they are computationally demanding and thus not considered here. To further analyze spectral features (π^* and $\pi + \sigma^*$), we calculated the density of states and partial polarizability of selected band transitions [10], respectively. Subsequently, Wannier function analysis, executed via Wannier90 [40], clarified the π electron orbitals and the π - π^* transitions based on maximally localized Wannier functions [35].

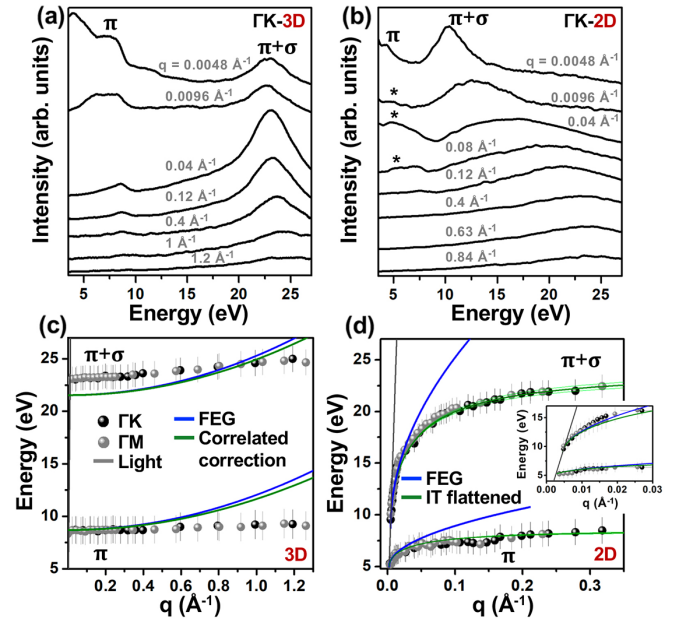


FIG. 2. (a), (b) the q -EELS spectra close to the light line at $q = 0.0048$ and $0.0096 \text{ \AA}^{-1}$ along ΓK [five-pixel integrations in Figs. 1(a) and 1(b)]. The spectra at $q = 0.04$ – $1.2 \text{ \AA}^{-1}$: those composing the respective Figs. 1(c) and 1(d). The asterisk denotes the weakly dispersive exciton. (c), (d) the superpositions of the respective pseudo-Voigt fitted plasmon peaks in 3D and 2D (black dots, ΓK ; gray dots, ΓM) with the light line (gray) and theoretical FEG (blue) and 3D-correlated-correction (green) and 2D-IT-flattened (green; IT, interband-transition) dispersions. Light-green curves in panel (d): the errors of $\pm 0.6 \text{ \AA}$ in the fitted ρ of $\sim 12 \text{ \AA}$. Inset in panel (d): the blowup showing the discrepancy from the FEG dispersion above $\sim 0.01 \text{ \AA}^{-1}$ (light-green curves, invisible in the figure scale). Error bars in panels (c) and (d) are same as in Fig. 1.

III. RESULTS AND DISCUSSION

Figure 1 maps the π - and $\pi + \sigma$ -plasmon dispersions in 3D and 2D MoS₂ along ΓK from the light line [Figs. 1(a) and 1(b)] to zone boundary at $1.322 \text{ \AA}^{-1}$ [Figs. 1(c) and 1(d)]. The pseudo-Voigt fitted peak positions of the plasmons are superimposed (Fig. S8 in the Supplemental Material [35]). Close to the light line, the intense spectral weights below the 3D π plasmon [Fig. 1(a)] are mainly contributed by excitonic excitations [32] and those in 2D [Fig. 1(b)] are also related to excitons [41] that are not the focus of this work and omitted hereinafter.

The 3D π and $\pi + \sigma$ plasmons are radiative inside the light cone [Fig. 1(a)], beyond which they become nonradiative and sharpened by the lifting of the radiative damping [Fig. 2(a)] [9]. In contrast, the 2D π and $\pi + \sigma$ plasmons are constantly outside the light cone and nonradiative [Fig. 1(b); Figs. S9, S10, and S15 in the Supplemental Material [35]]. Compared to the 3D plasmons [Fig. 2(a)], the 2D counterparts damp out quickly with increasing q and are noticeably diffused above $0.08 \text{ \AA}^{-1}$ [Fig. 2(b)], which, in effect, corresponds to $q_c^{2D} \sim 1/\rho$ and is to be cross-correlated with v_g/v_F later. Figures 2(c) and 2(d) outline the respective 3D- and 2D-plasmon dispersions, with those along ΓM (zone boundary, $1.145 \text{ \AA}^{-1}$) being also shown. Figures 2(c) and 2(d) show practically identical

TABLE I. The π and $\pi + \sigma$ electrons for the respective plasmons in 3D-bulk and 2D-ML MoS₂.

Electrons	Physical parameters	3D	2D
$\pi + \sigma$	Electron count per u.c.	18	18
	Electron density n	$3.36 \times 10^{23} \text{ (cm}^{-3}\text{)}$	$2.07 \times 10^{16} \text{ (cm}^{-2}\text{)}$
	Effective mass m^* (m_0)	1	0.5
	ε_∞	1	$1 + \rho q$
	Fermi velocity v_F (cm/s)	2.49×10^8	8.35×10^8
π	Electron count per u.c.	4	1
	Electron density n	$7.24 \times 10^{22} \text{ (cm}^{-3}\text{)}$	$1.15 \times 10^{15} \text{ (cm}^{-2}\text{)}$
	Effective mass m^* (m_0)	1	0.5
	ε_∞	1.32	$1 + \rho q$
	Fermi velocity v_F (cm/s)	1.49×10^8	1.97×10^8

ΓK and ΓM characteristics, revealing the isotropic ΓK and ΓM dispersions in both 3D [11] and 2D [25].

We quantitatively elaborate on the observed π - and $\pi + \sigma$ -plasmon dispersions in respective Figs. 2(c) and 2(d) within the RPA of FEG for 3D, $\omega_p^{3D}(\omega, q) = \omega_p^{3D} + \frac{3}{10} \frac{v_F^2 q^2}{\omega_p^{3D}}$, and 2D, ω_p^{2D} [1–3,14]. First, we show the calculated 3D $\pi + \sigma$ -plasmon dispersion in FEG [blue, Fig. 2(c)], for which the well-known parameters of n_{3D} , m^* , and ε_∞ are given in Table I [7,11]. More specifically, the 18 valence electrons of Mo and S in the 3D unit cell (u.c.) participate in the $\pi + \sigma$ plasmon and denote n_{3D} [7,11]. Using these parameters, the calculated theoretical $\hbar\omega_p^{3D}$ is 21.54 eV and v_F can be derived by $v_F = \frac{\hbar k_F}{m^*}$ (the Fermi wave vector $k_F = \sqrt[3]{3\pi^2 n_{3D}}$ in 3D) [10]. Noticeably, the theoretical $\hbar\omega_p^{3D}$ of the $\pi + \sigma$ plasmon is smaller than 23.06 eV in Fig. 2(c) and the q^2 dispersion in 3D FEG (blue) disagrees with the experiments [11]. We scrutinize this discrepancy by the Drude-Lorentz (DL) analysis, Eq. (1) [10], of the reported dielectric function of 3D MoS₂ at $q = 0.1 \text{ \AA}^{-1}$ [Fig. 3(a)] [8].

$$\varepsilon(\omega) = 1 - \sum_{l=1}^2 \frac{\omega_{p,l}^2}{\omega^2 + i\gamma_{p,l}\omega} - \sum_{l=1}^5 \frac{\omega_{T,l}^2}{\omega^2 - \omega_{T,l}^2 + i\gamma_{T,l}\omega}, \quad (1)$$

where the second term on the right is the Drude components (γ_p is the damping constant and l is the number of components) and the third term represents the Lorentz ones (ω_T is the single-particle onset and γ_T is the damping constant) [34], with Table S1 in the Supplemental Material detailing all parameters [35]. Figure 3(b) shows the DL-derived loss function $\text{Im}\{\frac{-1}{\varepsilon(\omega)}\}$ and its agreement with the reported one (dotted) and q -EELS at $q = 0.12 \text{ \AA}^{-1}$. The consistent DL depiction of $\varepsilon(\omega)$ in Figs. 3(a) and 3(b) requires the Drude π -plasmon ($\pi + \sigma$) component at 10 (14.7) eV [35], whereas the experimental excitation is observed at 8.7 (23.06) eV in Fig. 2(c). Unambiguously, the interband transition at ~ 11 eV in Fig. 3(a) (green) screens and pulls down (depolarizes and pushes up) the π ($\pi + \sigma$) plasmon [9].

Accordingly, the screened 3D π plasmon would be subject to an elevated ε_∞ of $(10/8.7)^2 \sim 1.32$ considering the $\sqrt{\frac{1}{\varepsilon_\infty}}$ dependence of ω_p^{3D} [10]. n_{3D} can then be derived (Table I),

corresponding to approximately four π electrons per u.c. principally of the S- p_z character (Fig. S17 in the Supplemental Material [35]). The reported six π electrons per u.c. in 3D, contributed by two Mo- d_z and four S- p_z electrons [7,42], are associated with the unawareness of the interband-transition impact and revisited herein. By this interband-transition effect, the 3D-FEG equation cannot capture the 3D π - and $\pi + \sigma$ -plasmon dispersions [blue, Fig. 2(c)], not even by incorporating the proposed correlated-correction factor

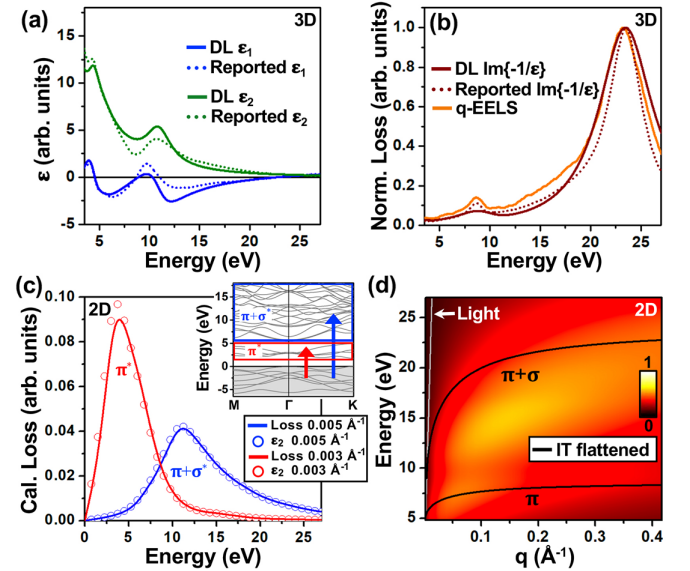


FIG. 3. (a) The DL analysis (solid lines) of the reported real- ε_1 and imaginary- ε_2 parts of 3D $\varepsilon(\omega)$ at $q = 0.1 \text{ \AA}^{-1}$ (dotted lines) [8]. (b) The calculated loss functions based on the DL and reported $\varepsilon(\omega)$, the former (latter) in solid (dotted) brown. Orange: the q -EELS spectrum of 3D MoS₂ [$q = 0.12 \text{ \AA}^{-1}$, Fig. 2(a)]. (c) The DFT-calculated loss functions for the respective π^* (red curve) and $\pi + \sigma^*$ (blue) interband transitions in ML MoS₂ at the designated light-line crossovers in q . Open dots: the corresponding imaginary- ε_2 part of $\varepsilon(\omega)$ and manifesting fermionic interband absorptions. Inset: the band structure with the π^* (red box) and $\pi + \sigma^*$ (blue box) bands accounting for the pertinent interband transitions (red and blue arrows). (d) The DFT loss functions of ML MoS₂ (color map) superimposed with the calculated IT-flattened dispersions of π and $\pi + \sigma$ plasmons (black curves) in Fig. 2(d). Gray: light line.

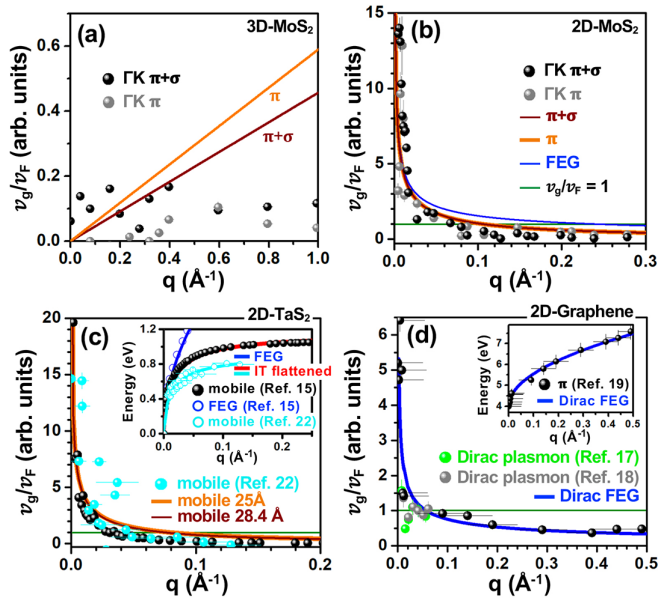


FIG. 4. (a) The theoretically (solid lines) and experimentally derived (dots, ΓK) v_g of the π and $\pi + \sigma$ plasmons in 3D MoS₂, scaled over the characteristic v_F . Part of the experimental data points omitted for clarity of the presentation. (b) The 2D theoretical (thin-brown, thick-orange, and thin-blue curves) and experimental (dots) counterparts. Error bars in panels (a) and (b) are same as in Fig. 1. (c) The v_g/v_F (solid-black and solid-cyan dots) derived from the reported IT-flattened dispersions of the mobile-charge plasmon in metallic ML TaS₂ [15,22]. The theoretical v_g/v_F (thick-orange and thin-brown curves), calculated by $m^* = 0.5 m_0$ [35] and respective ρ of 25 [15] and 28.4 Å [22] with the effect of the difference in ρ invisible in the figure scale. Inset: the reproduced dispersions (solid-black and open-cyan dots; open-blue dots, the FEG dispersion) [15,22]. Blue, red, and cyan curves (inset): the calculated dispersions using $m^* = 0.5 m_0$, $\rho = 25$ Å [15] (28.4 Å [22]), and fitted n_{2D} [35]. Error bars: those in Ref. [22]. (d) The v_g/v_F (light-green, gray, and black dots) derived from the reported dispersions of Dirac [17,18] and π [19] plasmons in graphene. The theoretical v_g/v_F (blue curve), calculated by $m^* = 0.06 m_0$ [18,19] and FEG $\epsilon_\infty = 1$. Inset: the reproduced gapped π -plasmon dispersion [19] fitted by quantum-mechanical ω_p^{QM} with $\omega_c = 4$ eV, $m^* = 0.06 m_0$, and $n_{2D} = 2.1 \times 10^{13} \text{ cm}^{-2}$ (blue curve) [35]. Error bars: those in Refs. [17–19]. Green line in panels (b)–(d): $v_g = v_F$.

of $[1 - \frac{1}{16}(\frac{\hbar\omega_p^{3D}}{E_F})^2]$ into the 3D-FEG equation of $\omega^{3D}(\omega, q)$ (green) [9].

Unlike the lack of a theoretical framework for the flattened 3D-plasmon dispersions [Fig. 2(c)], investigations on the mobile-charge plasmon in metallic ML TaS₂ pinpoint that the interband-transition flattened dispersion satisfies the non-local form in $\omega_p^{2D} = \sqrt{\frac{2\pi n_{2D} e^2}{m^*}} \sqrt{\frac{q}{\epsilon_\infty}}$ [inset, Fig. 4(c)] [15,22], where ϵ_∞ is formulated by the q -dependent Keldysh model of $\epsilon_\infty = \frac{1+\epsilon_s}{2} + \rho q$ (ϵ_s is the substrate dielectric constant) for $\rho q \ll \pi$ and $q_c^{2D} = \frac{1+\epsilon_s}{2\rho}$ [15,41,43]. The significance of q_c^{2D} has been proposed as the flattening of the 2D plasmon from the FEG dispersion ($\epsilon_\infty = 1$) [15]. Upon the reformulation of the Keldysh model into $\epsilon_\infty = 1 + q/q_c^{2D}$ ($\epsilon_s = 1$, the present freestanding case), we unravel the further

implication of q_c^{2D} , for which the electric-field lines connecting the moving charges in the nonlocal regime of $q > q_c^{2D}$ are largely localized in the 2D matter as characterized by $\epsilon_\infty > 2$ and growingly switch into vacuum at long wavelengths ($\epsilon_\infty \rightarrow 1$, $q < q_c^{2D}$). The nonlocal screening, considered general to ML TMD [15], is clearly associated with many-body electron interactions [44,45].

In ML MoS₂ [Fig. 2(d)], the 2D π and $\pi + \sigma$ plasmons are gapped, instead, as revealed by the respective light-line crossovers of $\sim 5.3 \pm 0.8$ and $\sim 9 \pm 0.5$ eV (Fig. S15 in the Supplemental Material [35]), which are consistent with the single-particle π^* and $\pi + \sigma^*$ interband transitions at the designated q 's in Fig. 3(c). Further nonlocal elaborations on the gapped π and $\pi + \sigma$ plasmons have, therefore, to incorporate the relevant fermionic term (ω_c) in order to convey the characteristic fermionic-to-bosonic turnover upon gaining q [19], resulting in $\omega_p^{2D} = \omega_c + \sqrt{\frac{2\pi n_{2D} e^2}{m^*}} \sqrt{\frac{q}{1+\rho q}}$ with the respective $\omega_c = cq_0$ (c is the speed of light and q_0 is the crossover at the light line) of ~ 5.3 and ~ 9 eV for the π and $\pi + \sigma$ plasmons.

In Fig. 2(d), we show the thus calculated dispersion of the $\pi + \sigma$ plasmon in nonlocal $\sqrt{\frac{q}{1+\rho q}}$ (green) and FEG $\sqrt{q}$ (blue, $\epsilon_\infty = 1$) using the n_{2D} integrated from the 18 valence electrons in the first Brillouin zone (Table I) [31], the reported m^* of $0.5 m_0$ in ML MoS₂ [41], and the only fitting parameter of ρ for the nonlocal dispersion. Upon the fitted ρ of $\sim 12 \pm 0.6$ Å, we capture the experimental $\pi + \sigma$ -plasmon dispersion up to $\sim 0.3 \text{ Å}^{-1}$ [green, Fig. 2(d)], beyond which the plasmon superimposes with the screening interband transition at ~ 21.9 eV (Figs. S8–S10 in the Supplemental Material [35]) and is significantly damped by this single-particle excitation. The corresponding ρq attains $\sim \pi$ ($\sim 2\pi$, ML TaS₂ [15]), substantially expanding the Keldysh model ($\rho q \ll \pi$) to large q , and the nonlocal screening holds in both the metallic and semiconducting ML TMD. The attempt to tackle the experimental π -plasmon dispersion using the same m^* and ρ [green, Fig. 2(d)] casts $n_{2D} \approx 1.15 \times 10^{15} \text{ cm}^{-2}$ (Table I), equivalent to one π electron per 2D u.c. of an S- p_z essence (Fig. S17 in the Supplemental Material), and this π -electron reduction from four in 3D is related to the lifted interlayer electronic hybridization in 2D [35]. The π electron in layered materials is entangled with the van der Waals force [7,29] and this 3D-to-2D reshuffling of the π -electron number, not reported before [24–28,41–43], suggests the 2D π -bonding weakening that is in line with the diminishing π -electronic hybridization in 2D [35].

Figure 3(d) exhibits the calculated plasmon-dispersion map in density-functional theory (without GW self-energy corrections) and satisfactorily captures the gapped, flattened dispersion of the 2D π plasmon (black). The omission of the full spectrum of GW renormalization effects nonetheless casts a discounted agreement with the 2D $\pi + \sigma$ plasmon [35], since the many-body corrections are essential in shifting quasiparticle bands [41,46] and high-energy optical transitions [47] to higher energies, the latter of which accounts for $\pi + \sigma^*$ [inset, Fig. 3(c)] associated with the $\pi + \sigma$ plasmon. The gapped, fermionic-to-bosonic characters of the 2D π and $\pi + \sigma$ plasmons are confirmed and the respective screening interband transitions are located at ~ 9 and ~ 21.9 eV

(Figs. S8–S10 in the Supplemental Material [35]), with the latter in agreement with an earlier report [31]. The key parameter of ρ for the nonlocal dispersions would be proportional to the electronic polarizability [43], underlining the larger $\rho \sim 25 \text{ \AA}$ in the 2D metal (ML TaS₂ [15]) than $\sim 12 \text{ \AA}$ in our 2D semiconductor.

Figure 4 exhibits our results on the q -dependent v_g , scaled over v_F to facilitate direct comparisons on the many-body, single-particle electron velocities. In Figs. 4(a) and 4(b) (black and gray dots), the experimental v_g is the first derivative of the characteristic dispersions [Figs. 2(c) and 2(d)] and then divided by the v_F in Table I. The theoretical v_g in 3D FEG is $v_g = \frac{\partial \omega^{3D}(\omega, q)}{\partial q} = \frac{3}{5} \frac{v_F^2 q}{\omega_p^{3D}}$, leading to $\frac{v_g}{v_F} = \frac{3v_F q}{5\omega_p^{3D}}$ [red and orange; Fig. 4(a)]. The experimental 2D counterparts are likewise calculated in Fig. 4(b) (dots), and the theoretical ones (curves) are derived as $\frac{v_g}{v_F} = \frac{e}{2\hbar} \sqrt{\frac{m^* g_v}{\epsilon_\infty q}}$ considering $k_F = \sqrt{\frac{4\pi n_{2D}}{g_s g_v}}$ in 2D for v_F , $g_s = 2$ (spin degenerate), and $g_v = 1$ (lifted valley degeneracy by the characteristic spin-orbit coupling [26]). For comparing to Fig. 4(b), the reported dispersions in ML TaS₂ [reproduced in Fig. 4(c), inset [15,22]] are also thus elaborated into v_g/v_F (cyan and black dots; curves, theoretical v_g/v_F with $m^* = 0.5m_0$ and associated n_{2D} [35]).

More specifically on the quantitative elaboration on the reported theoretical and experimental dispersions in ML TaS₂ [inset, Fig. 4(c)] [15,22], the similar band slopes in ML TaS₂ [$\sim 2.7 \text{ eV/\AA}$; Fig. 1(a) [15]] and ML MoS₂ [$\sim 2.8 \text{ eV/\AA}$; inset, Fig. 3(c)] near the Fermi level along ΓK or ΓM suggest that the m^* of $0.5m_0$ in ML MoS₂ [41] can be reasonably adopted, to the first approximations, to the ML TaS₂. Using the m^* of $0.5m_0$ and ω_p^{2D} equation in FEG ($\epsilon_\infty = 1$), the FEG plasmon dispersion in the freestanding ML TaS₂ in DFT is then captured by ω_p^{2D} with $n_{2D} \sim 2.4 \times 10^{14} \text{ cm}^{-2}$ [blue curve; inset, Fig. 4(c)]. Upon thus derived m^* and n_{2D} along with the reported ρ of $25 \text{ \AA}$ [15], the readily calculated IT-flattened dispersion ω_p^{2D} within the Keldysh model of $\epsilon_\infty(q) = 1 + 25q$ [red curve; inset, Fig. 4(c)] faithfully depicts the dispersion of the mobile-carrier plasmon in DFT (solid black dot) to $\sim 0.25 \text{ \AA}^{-1}$. Notably, the reported experimental plasmon dispersion in the ML TaS₂ [open cyan dots; inset, Fig. 4(c)] lies below the DFT counterpart (solid black dot), referring to a smaller n_{2D} in the experimental ML. Using m^* ($0.5m_0$) and reported ρ ($28.4 \text{ \AA}$ [22]), the experimental dispersion [open cyan dots; inset, Fig. 4(c)] is noticeably delineated by the IT-flattened dispersion in ω_p^{2D} with $n_{2D} \sim 1.7 \times 10^{14} \text{ cm}^{-2}$ (cyan curve), which corresponds to $\sim 70\%$ of the mobile charges derived from the above DFT dispersion ($n_{2D} \sim 2.4 \times 10^{14} \text{ cm}^{-2}$). To verify the consistency of thus estimated n_{2D} ($\sim 1.7 \times 10^{14} \text{ cm}^{-2}$) and also m^* ($0.5m_0$), it is useful to calculate the associated Drude weight of $\pi e^2 \frac{n_{2D}}{m^*}$ (in SI unit) reported in Ref. [22], where n_{2D} and m^* were treated as the single variable of Drude weight instead of analytical elaborations on individual n_{2D} and m^* like ours, and we obtain the corresponding Drude weight of $30 \times 10^{10} \Omega^{-1} \text{ s}^{-1}$ notably comparable to the reported $31.2 \pm 0.8 \times 10^{10} \Omega^{-1} \text{ s}^{-1}$ [22]. Unambiguously, our quantitative methodology for deriving n_{2D} , m^* , and ρ is robust. The lower experimental n_{2D} corresponding to Ref. [22] ($\sim 70\%$ of the theoretical counterpart [15]) would be an inherent property possibly ascribed to the

ML-TaS₂ sample preparation or so that is nonetheless beyond our scope.

Figure 4(d) shows the further comparison to graphene by thus elaborated v_g/v_F of the relativistic Dirac (light-green and gray dots [17,18]) and π (black dots [19]; reproduced, inset) plasmons, with the π -plasmon dispersion from the present research group's earlier work being incorporated into the inset (black dots) [19]. Characteristically, the relativistic fermions are of vanishing m^* ($\sim 0.06m_0$, π and Dirac electrons [18,19]) and feature the quantum-mechanical 2D-plasmon dispersion of ω_p^{QM} , with the characteristic degeneracy factor of $g = g_s g_v = 4$ owing to the fourfold degeneracy of graphene (the spin degeneracy $g_s = 2$; the valley degeneracy $g_v = 2$) [20]. Besides, the v_F in ω_p^{QM} is related to the vanishing m^* by $v_F = \frac{\hbar}{m^*} \sqrt{\frac{4\pi n_{2D}}{g_s g_v}}$ [21], which turns ω_p^{QM} into a function of n_{2D} and m^* as what we demonstrate above for the classical ω_p^{2D} . Using the reported $m^* \sim 0.06m_0$ [19]), the reported π -plasmon dispersion [solid black dots; inset, Fig. 4(d)] [19] is satisfactorily addressed by the ω_p^{QM} formulation with $n_{2D} \sim 2.1 \times 10^{13} \text{ cm}^{-2}$ in FEG (blue curve; $\epsilon_\infty = 1$). The derived n_{2D} of the π electrons is remarkably close to the n_{2D} of the Dirac fermions in doped graphene ($2 \times 10^{13} \text{ cm}^{-2}$ [17]; $1 \times 10^{13} \text{ cm}^{-2}$ [18]). For further deriving the theoretical v_g/v_F in graphene, one deals with the quantum-mechanical ω_p^{QM} and v_F [14], the latter of which is fundamentally identical to the classical $v_F = \frac{\hbar k_F}{m^*}$ as shown above [21]. We readily obtain the quantum-mechanical $\frac{v_g}{v_F} = \frac{e}{2\hbar} \sqrt{\frac{m^* g_v}{\epsilon_\infty q}}$ for graphene [blue curve, Fig. 4(d); $m^* = 0.06m_0$ [18,19,35]; $g_v = 2$ [20]]. Distinctly, the massive TMD and relativistic graphene show the same theoretical form of v_g/v_F , which merely depends on the microscopic parameters of m^* , g_v , and ϵ_∞ and is general to the mobile charges in metallic ML TaS₂, localized π and $\pi + \sigma$ electrons in semiconducting ML MoS₂, and relativistic fermions in graphene.

In 3D, the experiment-theory inconsistency [Fig. 4(a)] arises from the lack of a theoretical context incorporating the interband-transition effect [Fig. 2(c)], whereas it is clear that the 3D many-body electron motions are always slower than the single-particle v_F . In contrast, the π and $\pi + \sigma$ electrons in ML MoS₂ [Fig. 4(b)] manifest an elegant experiment-theory v_g/v_F agreement, confirming the sole dependence of the 2D-theoretical v_g/v_F on m^* , g_v , and ϵ_∞ . The concomitant collapsing of the experimental v_g/v_F of the π and $\pi + \sigma$ electrons [dots, Fig. 4(b)] into the same trend affirms the identical m^* and ϵ_∞ explored in Fig. 2(d) and Table I. Noticeably, the experimental v_g of the π and $\pi + \sigma$ plasmons in ML MoS₂ matches the v_F at $\sim 0.08 \text{ \AA}^{-1}$ [Fig. 4(b); Fig. S15 in the Supplemental Material [35]], which refers to $q_c^{2D} \sim 1/\rho \sim 0.08 \text{ \AA}^{-1}$ [15], and characterizes $v_g > v_F$ at $q < q_c^{2D}$. Moreover, the π and $\pi + \sigma$ plasmons become significantly damped above $q_c^{2D} \sim 0.08 \text{ \AA}^{-1}$ [Fig. 2(b) and Fig. S11 in the Supplemental Material [35]], for which the electric-field lines connecting the moving charges would be growingly confined in the 2D matter and the damping by associated single-particle excitations is to be most prominent at $q > 0.3 \text{ \AA}^{-1}$, irrelevant to q_c^{2D} . This characteristic reminds us of the accepted notion in thin plasmonic films that the thinner the film is, the more electric field is confined in the

substance, resulting in increased damping and reduced v_g of the plasmon by many-body electron interactions [44,45]. Such a many-body damping would address the damped plasmons in ML MoS₂ at $q < 0.3 \text{ \AA}^{-1}$. Meanwhile, the nonlocal dispersion in ML MoS₂ [green; inset, Fig. 2(d)] deviates from the FEG counterpart at $\sim 0.01 \text{ \AA}^{-1}$ (blue), at odds with the proposed 2D-plasmon flattening onset at $q_c^{2D} \sim 0.08 \text{ \AA}^{-1}$ [15]. We hereby propose q_c^{2D} as the prevailing many-body damping [45,46] with the experimentally observed concomitant $v_g = v_F$ [Fig. 4(b)], which sheds further light on Figs. 4(c) and 4(d).

The inspection on ML TaS₂ in Fig. 4(c) indeed unveils $v_g = v_F$ at $q_c^{2D} \sim 1/\rho \sim 0.04 \text{ \AA}^{-1}$ (ρ , $25 \text{ \AA}$ [15] or $28.4 \text{ \AA}$ [22]) and the experimentally observed plasmons are persistently broader than the DFT depiction without further correlations with the characteristic q_c^{2D} [22]. Although the origin for the plasmon broadening in ML TaS₂ remains unsettled [15,22], the above many-body damping shall be at work at the designated small q range [open cyan dots; inset, Fig. 4(c)]. Turning to graphene [Fig. 4(d)], the null interband-transition screening renders the FEG character of $\epsilon_\infty = 1$ (thus lifting ρ) [14,15], and both the relativistic Dirac and π electrons closely follow the same theoretical v_g/v_F (blue), as observed for the π and $\pi + \sigma$ plasmons in the massive ML MoS₂ [Fig. 4(b)]. While the absent ρ in graphene hinders a straightforward estimation of q_c^{2D} , the matching of $v_g = v_F$ is found at $q \sim 0.06 \text{ \AA}^{-1}$ [Fig. 4(d)] and might trigger the many-body damping [44,45], intriguingly coincident with the reported $q_c^{2D} \sim 0.06 \text{ \AA}^{-1}$ for damped Dirac plasmons, though, by the Landau damping [17,18].

IV. CONCLUSION

In summary, our experimental and theoretical elaborations on the q -dependent v_g in ML MoS₂ unravel the general scaling of $\frac{v_g}{v_F} = \frac{e}{2\hbar} \sqrt{\frac{m^* g_v}{\epsilon_\infty q}}$ for the massive ML MoS₂ and TaS₂ and the relativistic graphene. The q_c^{2D} experimentally concomitant

with $v_g = v_F$ would refer to the onset of many-body plasmon damping. Above q_c^{2D} , 2D plasmons could become slow ($v_g < v_F$) and damped, unlike the suggested robustness [15,22]. Persistent 2D plasmons of technological merits would be the long-wavelength ($q < q_c^{2D}$) ones and propagate at a distinctly higher speed than the single-particle motion ($v_g > v_F$). The v_g/v_F scaling and q_c^{2D} measure with the concomitantly observed $v_g = v_F$ await explorations across versatile 2D electron systems, in which the lighter m^* and faster v_F than those of the 3D counterpart exemplified in ML MoS₂ (Table I) and the fast many-body motion of $v_g > v_F$ [Fig. 4(b)] are all relevant to high-speed 2D electronics [23]. The q -EELS opens up vast opportunities for tackling emergent 2D phenomena.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [48].

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