Exotic surface plasmons in monolayer metal borophene

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The past decade has witnessed numerous discoveries in two-dimensional (2D) semimetals and insulators, including room temperature quantum Hall effect and topological states. On the contrary, metallic 2D materials have rarely been identified. Recently, borophene, a monolayer boron sheet, has emerged as a perfect 2D metal. Linear response calculations based on time-dependent density functional theory show that plasmons in borophene are strongly anisotropic with coexistence of gapless and gapped surface plasmon modes, which are originated from transitions between one-dimensional (1D) electron gas and Dirac electrons. The 1D plasmon is dissipationless, with energy dispersion extending far into the Brillouin zone boundary with large wavevectors ∼2 Å−1, supporting highly confined plasmon polaritons. The low-dimensional plasmons in intrinsic metal borophene open potential applications in plasmon waveguide and integrated optoelectronic devices at optical frequencies.

Many plasmonic devices, such as subwavelength waveguides, utilize thin metal films and/or metal-dielectric heterostructures [1]. Electromagnetic wave propagating along the interface thus couples with electron motion and forms surface plasmon polariton (SPP) [2, 3], which can be optically generated by prism geometry and/or nearfield optics. Noble metals (e.g. silver and gold) are traditionally used as free electron provider in the SPP devices [4–7]. However, propagation of SPP is limited by the dissipation in the metallic layer, bulk materials thus suffer from significant loss in SPP propagation [8]. Naturally, ultrathin two-dimensional (2D) materials, in particular graphene and related materials, are proposed to generate SPP with a low damping rate [9–23]. However, the main working regime of graphene plasmon is far-infrared, due to the low carrier density in semimetal graphene [24–30]. Other 2D materials, such as MoS2 [31–36] and phosphorene [37–40], are drawing great attentions thanks to their specific advantages such as the presence of hyperbolic and chiral plasmons. However, these materials are semiconductors with low electron densities, and are also limited to far-infrared to infrared working frequencies. Since the sources and detectors are less developed in infrared regime, 2D materials with higher electron density are extremely desirable for SPP devices working at optical frequencies.

Recently, borophene, the monolayer boron sheet, is experimentally synthesized via molecular beam epitaxy [41, 42]. Among several phases of borophene discovered, the β12 is particularly interesting because it is the most stable borophene polymorph and an intrinsic 2D metal [43, 44], differing from most other 2D materials. Angle-resolved photoemission spectroscopy (ARPES) have revealed the existence of Dirac cones in β12 and χ3 sheets [44, 45]. Moreover, theoretical calculations have demonstrated a variety of novel properties of borophene, such as universal scaling in resistivity and phonon-mediated superconductivity with a critical temperature ∼20 K [46, 47].

In this work we report a systematic study on plasmon excitation in the pristine borophene metal. Plasmon dispersion of the β12 borophene has been calculated using linear response (LR) time dependent density functional theory (TDDFT). Two branches of plasmons are observed: one is homogenenous along all directions, originating from collective excitations of bulk electrons in borophene. Surprisingly, the other plasmonic mode shows a strong anisotropic behavior, exhibiting exotic features such as visible light response (2-3 eV) in the large wavevector (1-2 Å−1) regime. The new plasmon mode at the visible range is attributed to transitions involving one-dimensional (1D) electron gas and Dirac electrons present in borophene electronic structure. This novel 1D plasmon mode makes borophene more suitable than graphene and phosphorene for anisotropic plasmon generation and integrated optoelectronics working at optical frequencies.

Results

Anisotropic surface plasmons. A boron atom has 3 valence electrons. In the two-dimensional limit, boron forms a hybrid lattice of hexagonal and triangle arrangements of atoms. The 2D lattice of borophene can be viewed as triangular lattice with periodic distributions of boron vacancies to stabilize its electronic structure. Different arrangements of these vacancies create various phases of borophene. The β12 structure is one of the most stable phases found in experiments [41, 42], which has been chosen as a representative structure in this study. Figure 1 shows the atomic structure of β12 borophene. The unit cell is rectangular with the side...
FIG. 1. (a) The structure and (b) Brillouin zone of $\beta_{12}$ borophene. (c-d) EELS along $\Gamma$-X and $\Gamma$-Y directions shown in (b). The white and yellow shaded zones in (c) represent regimes for pair-like excitations of the 1D and bulk electrons, respectively.

length $a_1 = 2.92$ Å and $a_2 = 5.06$ Å, consisting of five boron atoms. The vacancies line up along the horizontal direction (defined as X direction). This special structure would introduce anisotropy between the horizontal (X) and vertical (Y) directions in both real and momentum space.

In general, the anisotropy in lattice structures could potentially lead to anisotropy in the electronic structure and collective excitation. To analyze such anisotropic features of surface plasmons in borophene, we calculate electron energy loss spectroscopy (EELS) along the $\Gamma$-X and $\Gamma$-Y directions, as shown in Fig. 1. Two clear plasmon branches are observed. One branch locates at 3 eV and increase rapidly with the wavevector $q$ of plasmon. This branch shows homogeneous energy dispersions $E(q)$ along all directions. Thus, we label it as $h$ mode. Another branch locates at a lower energy than the $h$ mode and shows anisotropic dispersion along different directions. Along $\Gamma$-X, this branch starts from 0 eV at $q = 0$ and increases to the maximum at the half of the reciprocal lattice vector $q = |b_1|/2 = 1.07$ Å$^{-1}$, then decreases to 0 at $q = |b_1| = 2.14$ Å$^{-1}$ (Fig. 2). Along $\Gamma$-Y, this branch at small $q$ is upward shifted leaving an excitation gap of 1.9 eV, but is suppressed in its energy spreading range. Due to its anisotropic behavior, we name this branch as $a$ mode.

The strong anisotropic plasmon observed in borophene is different from other 2D materials such as graphene and phosphorene. At low energies, the plasmon branches in phosphorene behave like 2D plasmons with an energy difference of only 0.3 eV along $\Gamma$-X and $\Gamma$-Y direction [38]. The small anisotropy is also reported in graphene [24] and other phases of borophene [55], which was attributed to small differences in orientation-dependent dispersion of 2D electron gas. In contrast, for $\beta_{12}$ borophene, the plasmon is highly distinguishable between $\Gamma$-X and $\Gamma$-Y directions. The plasmon at the low energy range (0-2 eV) can be only excited along $\Gamma$-X. We note that the highly anisotropic plasmon in $\beta_{12}$ borophene is not a trivial consequence of the rectangle lattice. Further analysis of the electronic structure of borophene is needed to reveal the origin of the anisotropy in plasmon excitation.

We note that the low-energy plasmon mode shows strong responses in a very broad energy range from far-infrared ($<1.0$ eV), to visible and ultraviolet light (up to 3.2 eV) region (Fig. 2). In addition, the corresponding wavevector for plasmon excitation in borophene is very large, reaching $\sim 2$ Å$^{-1}$ without a significant Lautau damping loss. Thus extreme confinement of polariton propagation with a short wavelength $\sim 3$ Å can be reached.
FIG. 2. The EELS spectrum at different $q$ along $\Gamma$-X and $\Gamma$-Y. The $q$ values are starting from 0.015 Å to 2.14 Å (1.20 Å) along $\Gamma$-X ($\Gamma$-Y). The curves are shifted downwards in vertical axis for a better illustration.

Electronic structure and spectra decomposition. The electronic structure of $\beta_{12}$ borophene is shown in Fig. 3. The Fermi surface of borophene consists of three parts: (I) the ribbon along $\Gamma$-Y direction centered at X point, resulting from the 1D electron gas (1DEG) dispersed along $\Gamma$-X; (II) the ring centered at Y point, featuring bulk electrons; (III) the small hole pockets at $\Gamma$ point. Among these three parts, only the first term, i.e. the 1DEG is strongly anisotropic. However, the 1D plasmon dispersion $E(q)$ was found to increase monotonously with $q$, in both theoretical modeling [56, 57] and the experimental observation in atomic metal chains [58–60]. While in borophene, the plasmon formed by 1D electron gas has a hat-like energy dispersion as shown in Fig. 1 and Fig. 2. We also note that above the Fermi level, there exist two types of Dirac cones on the $\Gamma$-X and Y-S direction, respectively, in agreement with experimental measurements [44]. Since the Dirac point is $\sim$2 eV above the Fermi level, borophene can be considered as the extremely hole-doped graphene in some sense.

To make connections between plasmon dispersion in the energy loss spectrum and the electronic band structure, we analyze the contributions from different electron-hole pairs to the EELS spectrum in the independent particle (IP) approximation. As shown in Fig. 4, the spectrum at the IP level is similar to that calculated by RPA, but the peaks are generally higher in RPA spectra. At small $q$, the difference between RPA and IP is large, since the interaction kernel $K_{G_1 G_2}(q)$ is divergent at small $q$. With $q$ increases, the IP and RPA spectrum become closer to each other. At 2.06 Å$^{-1}$, the EELS calculated with these two approximations are almost identical. The peaks of the anisotropic branch can be distinguished in the loss spectrum calculated by IP, which locates at 0.5, 1.2, 0.5 eV for $q = 0.09, 0.36, 2.06$ Å$^{-1}$, respectively. The anisotropic branch is mainly contributed by the intraband transition in band No. 7, which crosses the Fermi level (Fig. 3).

To visualize the excitation mode of this plamon in $q$ space, we show the contour plot of the energy difference $\omega_{n,n'}(q,k)$ between the initial $\{n,k-q\}$ and final state
FIG. 3. Electronic band structure of $\beta_{12}$ borophene in (a) three-dimensional plot and (b) conventional two-dimensional plot.

\[ \{n', k\} \text{ as a function of } k, \text{ defined by} \]

\[ \omega_{n,n'}(q, k) = \begin{cases} 
\epsilon_{n'k} - \epsilon_{nk-q} & \epsilon_{n'k} > 0 \cap \epsilon_{nk-q} < 0 \\
0 & \text{else} \end{cases}, \]

where $\epsilon_{nk}$ is the eigenvalue of band $n$ at $k$ point. As shown in Fig. 4(d-f), the major excitation mode at $q = 0.09 \text{ Å}^{-1}$ comes from the oscillation of electrons in the 1DEG. At $q = 0.36 \text{ Å}^{-1}$, the plasmon is mainly contributed by the excitation from 1DEG to the unoccupied states around the Dirac point, shown in the energy contour at the IP peak of 1.2 eV. At large $q$ with $q = 2.06 \text{ Å}^{-1}$, electronic transitions between two 1DEG at $-X$ and $X$ are observed. Thus, the excitation mode of the anisotropic plasmon branch is mainly contributed by the intraband excitation of the 1DEG and Dirac electrons.

**One-dimensional electron gas model.** To describe the behavior of 1D electrons in borophene, we adopt a free electron gas model [57, 61, 62], which is widely used in describing plasmons of atomic chains [58–60, 63–65]. Since the model describes the plasmon of 1DEG at $\Gamma$ point (1DEG@\Gamma, dashed green line), the 1D electron gas centered at X point (1DEG@X) in borophene is treated as a 1DEG@\Gamma with additional excitation channels. For free electron gas, $\omega_k = k^2/2m$ (assuming $\hbar = 1$ and $m$ is the free particle mass). For any given $q$, we have the conventional excitation channel:

\[ \omega_{+,k,q} = \omega_{k+q} - \omega_k = (kq + \frac{1}{2}q^2)/m, \]

and a new channel:

\[ \omega_{-,k,q} = \omega_{k+q} - \omega_k \approx \omega_{k-q} - \omega_k = (-kq + \frac{1}{2}q^2)/m. \]

In above analysis the plus sign originates from $k$ to $k+q$ excitation denoted by green arrow in Fig. 5, which is well described in conventional 1DEG plasmon models [57, 58], while the minus sign is contributed by $-k$ to $-k+q$ excitation denoted by the blue arrow ($k \approx X$), which is the new excitation channel due to the unique 1DEG in borophene centered at the edge of first Brillouin zone.

The condition for the emergence of plasmon excitation is

\[ \epsilon(q, \omega_p) = 1 - V(q)\text{Re}\chi_0(q, \omega_p) = 0, \]

where $\epsilon$ is the dielectric function, $\omega_p$ is the frequency of plasmon, $V(q)$ is Coulomb potential and $\chi_0$ is the IP response function [56]:

\[ \chi_0(q, \omega_p) = \frac{1}{\pi} \int dk \theta(q \pm k - k_F)\theta(k_F \mp k) \left[ \frac{1}{\omega - \omega_{\pm,k,q} + i\eta} - \frac{1}{\omega + \omega_{\pm,k,q} + i\eta} \right]. \]

Therefore

\[ \text{Re}\chi_0(q, \omega_p) = \pm \frac{1}{\pi q} \ln \frac{\omega^2 - \omega_+^2}{\omega^2 - \omega_-^2}, \]
FIG. 4. (a-c) The EELS at different $q$ with independent particle approximation. The black dash lines denote the peaks of the anisotropic branch at different $q$, which locate at 0.5 eV, 1.23 eV, 0.5 eV, respectively. The transition $i \rightarrow j$ denotes the EELS contribution from band $i$ to band $j$, as labeled in Fig. 3. The brown dash line is the EELS with RPA for comparison. (d-f) The contour of $\omega_7,\omega_7(q,k) = \epsilon_{7,k} - \epsilon_{7,k-q}$ at different $q$. The arrows denote the excitation mode connecting states at $k-q$ and $k$.

where

$$\omega^2_\pm = \left| k_F q \pm \frac{1}{2} q^2 \right| / m, \quad (7)$$

is the upper and lower limit of the pair-like excitation regime. Combining Eq. (6) with Eq. (4), we get the two solutions of plasmon dispersion of 1DEG as

$$\omega_p(q) = \left[ \frac{A(q) \omega^2_+ - \omega^2_-}{A(q) - 1} \right]^{\frac{1}{2}}, \quad (8)$$

and

$$\omega_p(q) = \left[ \frac{A(q) \omega^2_- - \omega^2_+}{A(q) - 1} \right]^{\frac{1}{2}}. \quad (9)$$

Here

$$A(q) = \exp \left( \frac{\pi q}{m V(q)} \right). \quad (10)$$

$V(q)$ is Coulomb potential

$$V(q) = \frac{e^2}{K} b^2 q^2 \int_0^\infty \frac{e^{-x}}{x} dx, \quad (11)$$

where $K$ is the dielectric constant, $b$ is the width of the 1DEG.

Using parameters listed in Table I, this model yields the result consistent with the TDDFT calculations, as shown in Fig. 5(c). Clearly, the hat-like plasmon comes from Eq. (9). We note that, among the parameters listed in Table I, $k_F$ is the length of $\Gamma$-$X$ and not adjustable. The $b$ and $K$ have negligible effect on $\omega_p(q)$ when $q > 0.1 \ \text{Å}^{-1}$. The $m$ is the effective mass of electrons in solid, which could be obtained by fitting the band structure of 1DEG (the green dash line in Fig. 5(b)). Here, the mass $m = 2.4m_0$ used in Eq. (10) is slightly larger than the effective mass evaluated from the band structure $m^* = 1.6m_0$. It justifies the electron gas model to describe electrons in borophene.

We give a brief discussion about the upper-energy-limit and lower-energy-limit plasmon branches. With $q \rightarrow \infty$, 

| $k_F$ (Å$^{-1}$) | $b$ (Å) | $K$ | $m$ |
|----------------|----------|-----|-----|
| 1.07 Å$^{-1}$  | 4 Å      | 6.25 | 2.41$m_0$ |
FIG. 5. (a) Fermi surface of borophene. The black solid rectangle represents the reciprocal lattice. (b) The band structures along X-Γ-X. Fermi energy is set as zero. The green dash line follows $E_k = k^2/2m^* + E_0$ with $E_0 = -3.26$ eV and $m^* = 1.6m_0$, representing the band of 1DEG@Γ with the same dispersion with that of 1DEG@X at $k > 1.07 \text{Å}$. The green arrow represents the excitation channel of 1DEG@Γ. The blue arrow represents the new excitation channel of 1DEG@X. (c) The plasmon dispersion calculated by the 1DEG model. The empty circles represent the TDDFT data and the solid line represent data predicted by the model in Eq. (9).

Discussion

We have studied the plasmon excitation of borophene and identified a novel plasmon mode, which is originated from the unique 1D electron gas present in borophene band structure. Absent in other 2D materials, we observe the coexistence and interplay of the 1D plasmon mode and conventional plasmon excitation, which have not been reported before. Different from the conventional 3D, 2D and 1D plasmonic mode discussed in literature, the new 1D plasmon excitation exhibits unusual and desirable properties such as the parabolic-like energy dispersion, strong response in visible light range, and surviving in large wavevector regime. Based on the electronic structure, an analytic model analysis reveals that the novel 1D plasmon comes from collective intraband excitations of 1DEG and Dirac electrons across the Fermi level. The exotic features such as visible light response (2-3 eV) and extreme large wavevector (1-2 Å$^{-1}$) of the new plasmon excitation make borophene a promising candidate for applications in nanophotonics and integrated optoelectronics working at optical frequencies.

Methods

Linear response theory for plasmon excitations.

The frequency and wave-vector dependent density response functions are calculated within time-dependent density-functional theory (TDDFT) formalism using the random phase approximation for exchange-correlation functional. The non-interacting density response function in real space is written as

$$\chi^0(r, r', \omega) = \sum_{k,n,n'} \sum_{k,q} \sum_{\omega} \frac{f_{nk} - f_{nk+q}}{\omega + \epsilon_{nk} - \epsilon_{nk+q} + i\eta} \times \psi^*_{nk}(r)\psi_{nk+k+q}(r') \psi^*_{nk+k+q}(r')$$

where $\epsilon_{nk}$ and $\psi_{nk}(r)$ are the eigenvalues and eigenvecto-
tors of the ground state Hamiltonian. For translation invariant systems, $\chi^0$ can be expanded in plane-wave basis as

$$\chi^0(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{\Omega} \sum_{\mathbf{G}, \mathbf{G}'} e^{i(\mathbf{q}+\mathbf{G}) \cdot \mathbf{r}} \chi^0_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega) e^{-i(\mathbf{q}+\mathbf{G}') \cdot \mathbf{r}'},$$

where $\Omega$ is the normalization volume, $\mathbf{q}$ stands for the Bloch vector of the incident wave and $\mathbf{G}$($\mathbf{G}'$) are reciprocal lattice vectors. The full interacting density response function is obtained by solving the Dyson’s equation, from its non-interacting counterpart $\chi^0$ as

$$\chi(\mathbf{r}, \mathbf{r}', \omega) = \chi^0(\mathbf{r}, \mathbf{r}', \omega) + \int \int_\Omega d\mathbf{r}_1 d\mathbf{r}_2 \chi^0(\mathbf{r}, \mathbf{r}_1, \omega) K(\mathbf{r}_1, \mathbf{r}_2) \chi(\mathbf{r}_2, \mathbf{r}', \omega),$$

where the kernel is the summation of coulomb and exchange-correlation (XC) interaction

$$K(\mathbf{r}_1, \mathbf{r}_2) = \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} + f_{xc},$$

Here, $f_{xc} = \partial V_{xc}[n]/\partial n$ is the XC kernel. The common used XC kernels include adiabatic local density approximation [66], Bootstrap approximation [67], etc. A simplest case is the so-called random phase approximation (RPA), with $f_{xc} = 0$. Since the plasmon is demonstrated to be well describe in RPA [68], we use RPA in the following calculation, while some results with other XC kernels are also tested and found to be consistent with RPA results. With translational symmetry, it is more convenient to represent $\chi^0$ in the reciprocal lattice space. Fourier coefficients $\chi^0_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega)$ are written as

$$\chi^0_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega) = \sum_{n, n'} \chi^0_{\mathbf{G}\mathbf{G}', n, n'}(\mathbf{q}, \omega),$$

where

$$\chi^0_{\mathbf{G}\mathbf{G}', n, n'}(\mathbf{q}, \omega) = \frac{1}{\Omega} \sum_{\mathbf{k}} \frac{f_{nk} - f_{n'k+\mathbf{q}}}{\omega + \epsilon_{nk} - \epsilon_{n'k+\mathbf{q}} + i\eta} \langle \psi_{nk}|e^{-i(\mathbf{q}+\mathbf{G}) \cdot \mathbf{r}} \psi_{n'k+\mathbf{q}}\rangle_{\text{cell}} \langle \psi_{nk}|e^{i(\mathbf{q}+\mathbf{G}') \cdot \mathbf{r}' \psi_{n'k+\mathbf{q}}\rangle_{\text{cell}},$$

and so is the the Dyson’s equation

$$\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega) = \chi^0_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega) + \sum_{\mathbf{G}_1, \mathbf{G}_2} \chi^0_{\mathbf{G}_1\mathbf{G}_2}(\mathbf{q}, \omega) K_{\mathbf{G}_1\mathbf{G}_2}(\mathbf{q}) \chi_{\mathbf{G}_2\mathbf{G}'}(\mathbf{q}, \omega).$$

(18)

The dielectric function can be expressed with $\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega)$ as

$$\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega) = 4\pi \frac{\delta_{\mathbf{G}\mathbf{G}'} - \sum_{\mathbf{G}_1} K_{\mathbf{G}_1\mathbf{G}}(\mathbf{q}) \chi_{\mathbf{G}_1\mathbf{G}'}(\mathbf{q}, \omega).}{\Omega}$$

(19)

Within RPA, the coulomb kernel becomes diagonal in reciprocal space

$$K_{\mathbf{G}\mathbf{G}'}(\mathbf{q}) = \frac{4\pi}{|\mathbf{q} + \mathbf{G}|^2} \delta_{\mathbf{G}\mathbf{G}'}, \delta_{\mathbf{G}\mathbf{G}'}.$$  

(20)

Thus, the dielectric function is simplified as

$$\epsilon^{-1}_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega) = \delta_{\mathbf{G}\mathbf{G}'} - \frac{4\pi}{|\mathbf{q} + \mathbf{G}|^2} \chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega).$$

(21)

The macroscopic dielectric function is defined by

$$\epsilon_M(\mathbf{q}, \omega) = \frac{1}{\epsilon_{00}(\mathbf{q}, \omega)},$$

(22)

and the electron energy loss spectrum (EELS) is

$$\text{EELS} = -\text{Im} \frac{1}{\epsilon_M(\mathbf{q}, \omega)}.$$  

(23)

For comparison and analysis, simpler approximation called independent particle (IP) is occupied. By setting $K_{\mathbf{G}_1\mathbf{G}_2}(\mathbf{q}) = 0$ in Eq. 18,

$$\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega) = \chi^0_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega).$$

(24)

With IP, the EELS spectra can be calculated as

$$\text{EELS} = \sum_{nn'} \epsilon^{-1}_{nn'}(\mathbf{q}, \omega),$$

(25)

where

$$\epsilon_{nn'}(\mathbf{q}, \omega) = \frac{1}{\epsilon_{00,nn'}(\mathbf{q}, \omega)} = \frac{1}{1 - \frac{4\pi}{|\mathbf{q} + \mathbf{G}|^2} \chi^0_{00,nn'}(\mathbf{q}, \omega)},$$

(27)

It means that, the EELS($\omega$) is solely determined by $\chi^0_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega)$, which is only contributed by transition from band $n$ and to $n'$ satisfying $\epsilon_{nk+\mathbf{q}} - \epsilon_{nk} = \omega$. Furthermore, since the contribution of $\chi^0_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \omega)$ to $\chi^0$ is independent, the EELS can be decomposed to EELS$_{nn'}$, corresponding to the EELS contribution from $n \rightarrow n'$. 

**Time-dependent density functional theory calculations.** The frequency and wavevector dependent density response functions are calculated within the LR-TDDFT formalism using random phase approximation as implemented in the GPAW package [48–52]. The projector augmented-waves method and Perdew-Burke-Ernzerhof exchange-correlation [53] are used for the ground state calculations. The plane-wave cutoff energy is set to be 500 eV. The thickness of vacuum layer is set to be larger than 10 Å. The Brillouin zone is sampled using the Monkhorst-Pack scheme [54] with a dense $k$-point mesh $142 \times 72 \times 1$ in the self-consistent calculations.
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