

Substrate screening approach for quasiparticle energies of two-dimensional interfaces with lattice mismatch

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Two-dimensional (2D) materials are outstanding platforms for exotic physics and emerging applications by forming interfaces. In order to efficiently take into account the substrate screening in the quasiparticle energies of 2D materials, several theoretical methods have been proposed previously but are only applicable to interfaces of two systems' lattice constants with certain integer proportion, which often requires a few percent of strain. In this work, we analytically showed the equivalence and distinction among different approximate methods for substrate dielectric matrices. We evaluated the accuracy of these methods by applying them to calculate quasiparticle energies of hexagonal boron nitride interface systems (heterojunctions and bilayers) and compared the results with explicit interface calculations. Most importantly, we developed an efficient and accurate interpolation technique for dielectric matrices that made quasiparticle energy calculations possible for arbitrarily mismatched interfaces free of strain, which is extremely valuable for practical applications.

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I. INTRODUCTION

Two-dimensional (2D) materials and their interfaces have shown unprecedented rich physics and promising applications in many areas, such as optospintronics [1,2], quantum information [3,4], and biomedical research [5,6]. Emerging phenomena such as nonconventional superconductivity [7,8] and topologically protected states [9–12] may be created by stacking 2D layers. Experimentally, growth of 2D materials, achieved through physical epitaxy or chemical vapor deposition (CVD), is typically supported on a substrate [13]. In general, the electrical and optical properties of 2D materials could be strongly modified by substrate screening. For example, the 2D materials' fundamental electronic gap can be significantly reduced due to the dielectric screening from surrounding layers (substrates) when forming heterointerfaces [14,15]. Reliable prediction of substrate screening effects from first-principles calculations is critical for accurate interpretation of experimental results and guidance for design of new materials.

Currently, widely used electronic structure methods such as the HSE06 hybrid functional [16] may accurately describe a large number of three-dimensional bulk systems but are inadequate for low-dimensional systems such as ultrathin 2D materials because of their highly inhomogeneous dielectric screening. The Koopman's compliant hybrid functional [17–19] or dielectric-dependent hybrid functional [20] are necessary for the electronic structure of ultrathin 2D materials, where the fraction of Fock exchange α varies with the number of layers [21] and needs to be determined for each individual material and thickness.

On the other hand, many-body perturbation theory (MBPT) [22–24] can successfully describe the quasiparticle properties of 2D materials such as fundamental electronic gaps, regardless of their thickness and dielectric properties. Generally, one- and two-particle excitations, experimentally corresponding to charged excitations (e.g., photoemission) and neutral excitation (e.g., optical absorption), can be accurately obtained by the GW approximation [22,25–28] and the Bethe-Salpeter equation [29–34] (BSE), respectively. However, explicit interface calculations at this level of theory are extremely computationally demanding and not suitable for the rapid evaluation of the effect of different substrates.

Therefore, several approximate methods have been proposed to compute the quasiparticle properties of interfaces at the cost of primitive cell calculations of the subsystems composing the interface [15,35,36]. Typically, for weakly bonded van de Waals (vdW) interfaces, the hybridization between layers is relatively weak and the dominant effect of the substrate consists in modifying the dielectric screening of the material of interest [36]. Within the GW approximation, this effect can be described by approximating the dielectric matrix of an interface in terms of contributions from individual subsystems (the material and the substrate), as proposed in several previous studies [15,35,36]. Despite the reasonable level of accuracy achieved through these methods, the underlying approximations and connections between different methods have not been carefully evaluated. For example, partially neglecting the local-field effect of substrate dielectric screening (i.e., removing in-plane and/or out-of-plane off-diagonal elements of dielectric matrices [15,37,38]) has been a common approximation, which was not carefully examined before. We will test the applicability of such approximation in different systems for both in-plane and out-of-plane components of dielectric matrices.

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Most important, previous methods cannot be applied to arbitrarily lattice-mismatched 2D interfaces; namely, an integer relation between lattice constants is necessary ($L \cdot N = \tilde{L} \cdot \tilde{N}$, where L and $\tilde{L}$ are the primitive lattice constants of the two systems at interfaces and $N(\tilde{N})$ is an integer number). Forcing lattice matching or the fulfillment of the above relation is typically required for interface calculations. These constraints either limit the choice of interfaces that can be studied or require applying artificial strain that may strongly modify the electronic structure. In this work, we develop a reciprocal-space linear-interpolation method in the entire $\mathbf{q} + \mathbf{G}$ space to approximate interface dielectric matrices of arbitrarily mismatched systems. This approach makes MBPT calculations of general interfaces possible and free of strain.

In order to demonstrate the accuracy and efficiency of this methodology, we will consider applications to interfaces involving hexagonal boron nitride (hBN). This material has a wide band gap in ultraviolet region, with promising applications in deep ultraviolet light-emitting devices [39] and as a host for spin qubits and single photon emitters [40] in quantum information technologies [26,41]. As ultrathin hBN is mostly supported on substrates in experimental measurements, it is critical to accurately predict the effect of substrates on electronic structure of hBN. This is also important for evaluations of defect properties in 2D materials supported by substrates [42,43]. We will use hBN with SnS_2 substrates and bilayer hBN in two conformations as prototypical examples for our methodology validation in this study as shown in Fig. 1.

For the rest of the paper, we first analytically derived the connection among different approximations of dielectric matrices with substrate screening [15,36,38,44,45]. We then performed the separate GW calculations for subsystems from interfaces with several approximate approaches to construct the interface polarizability, and compared results with explicit interfaces in order to evaluate the accuracy of these methods. Next we examined the importance of off-diagonal elements of polarizability in substrate screenings in various 2D interface systems. Finally, we introduced our linear-interpolation technique, benchmarked it, and showed the quasiparticle energies obtained by this technique for arbitrarily lattice-mismatched 2D interfaces.

II. METHODOLOGY

In this section, we will discuss the different methods and concepts used in this paper, which are summarized in Table I.

A. Methods for interface polarizability

The interactions among quasiparticles within the GW approximation are described by the screened Coulomb potential $W = \epsilon^{-1}v_C$, where v_C is the bare Coulomb interaction and ϵ is the dielectric matrix. The inverse dielectric matrix is defined by $\epsilon^{-1} = \mathbb{1} + v_C\chi$ within the random-phase approximation (RPA) [23]. The reducible polarizability χ can be obtained from the irreducible polarizability χ_0 (also known as independent-particle polarizability) through the equation $\chi = \chi_0 + \chi_0 v_C \chi$.

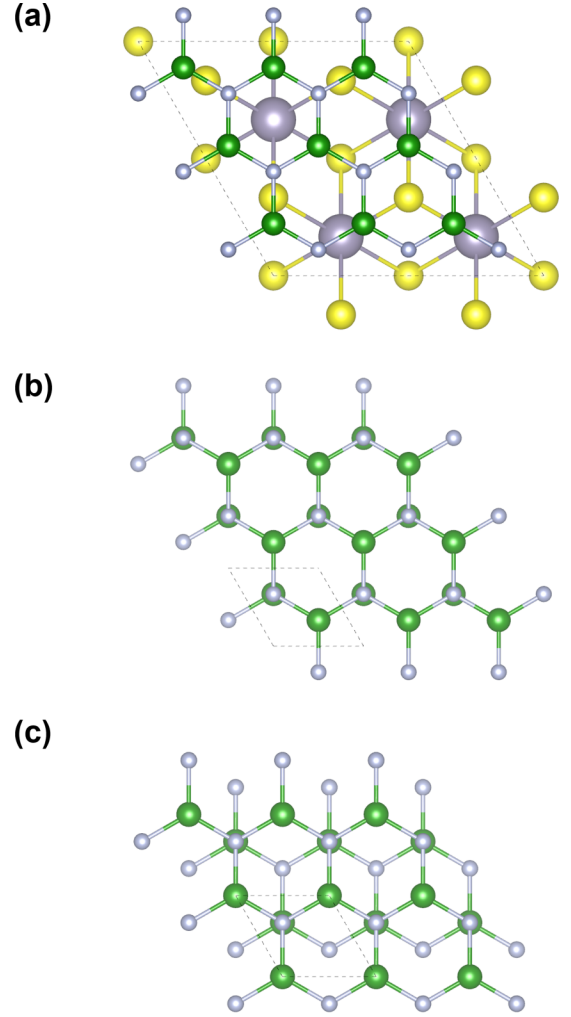


FIG. 1. Atomic structures of 2D interfaces (a) hBN/ SnS_2 , (b) bilayer hBN with AB stacking, and (c) bilayer hBN with AA' stacking. The green balls denote boron atoms; the white balls denote N atoms; the yellow balls denote sulfur atoms; and the silver balls denote Sn atoms.

TABLE I. Overview of methodology in this work.

Methods	Assumption
$\chi_{\text{eff}}^{\text{sum}}$ [Eq. (3)]	Coulomb interaction between layers
$\chi_{\text{eff}}^{\text{GSC}}\text{-sum}$	Uses interface eigenvalue in $\chi_{\text{eff}}^{\text{sum}}$
$\chi_{\text{eff}}^{\text{FWF}}\text{-sum}$	Uses interface eigenvalue and wave functions in GW with $\chi_{\text{eff}}^{\text{sum}}$
$\chi_0\text{-sum}$ [Eq. (4)]	Coulomb interaction between layers, equiv. to $\chi_{\text{eff}}^{\text{sum}}$ at RPA
$\chi\text{-sum}$ [Eq. (6)]	No interaction between layers
Approximations	Definition
$\epsilon^{-1}\text{-diag}$	Neglects χ^s off-diagonal elements
$\epsilon\text{-diag}$	Neglects χ_0^s off-diagonal elements
Interface structure	Solution
Lattice match	Direct summation
Special match	$\mathbf{q} + \mathbf{G}$ mapping
Arbitrary mismatch	$\mathbf{q} + \mathbf{G}$ bilinear interpolation

For the purpose of our discussion, we first partition the total (“tot”) vdW interface systems into material (“m”) and substrate (“s”) subsystems [36]. Considering density response of external field, we obtain

$$\begin{aligned}\delta n^m &= \chi^m(\delta V_{\text{ext}} + v_C \delta n^s), \\ \delta n^s &= \chi^s(\delta V_{\text{ext}} + v_C \delta n^m),\end{aligned}\quad (1)$$

where δn is the density response and the reducible polarizability χ is defined as the density-density response function to an applied potential. If we consider the material subsystem (“m”) as the probe, the total external potential includes the external applied potential (δV_{ext}) and the Coulomb potential from the charge response δn^s in the substrate ($v_C \delta n^s$). (We assume the material and substrate are connected only through interlayer Coulomb interactions, with minimum wave-function overlap between material and substrates, i.e., interlayer hybridization.) Then we define an effective polarizability χ_{eff} as a density response function of one subsystem to only the external applied potential (δV_{ext}), i.e., $\chi_{\text{eff}}^{m/s} = \delta n^{m/s} / \delta V_{\text{ext}}$. More precisely, $\chi_{\text{eff}}^{m/s}$ can be given in terms of $\chi^{m/s}$ through Eq. (1):

$$\begin{aligned}\chi_{\text{eff}}^{m/s} &= \frac{\delta n^{m/s}}{\delta V_{\text{ext}}} \\ &= (1 - \chi^{m/s} v_C \chi^{s/m} v_C)^{-1} (\chi^{m/s} + \chi^{m/s} v_C \chi^{s/m}).\end{aligned}\quad (2)$$

When subsystems have negligible interlayer wave-function overlap (i.e., hybridization), the total density response (δn^{tot}) can be written as $\delta n^{\text{tot}} = \delta n^m + \delta n^s$ and then the total polarizability χ^{tot} of entire interface systems is

$$\chi^{\text{tot}} = \frac{\delta n^{\text{tot}}}{\delta V_{\text{ext}}} = \frac{\delta n^m}{\delta V_{\text{ext}}} + \frac{\delta n^s}{\delta V_{\text{ext}}} = \chi_{\text{eff}}^m + \chi_{\text{eff}}^s. \quad (3)$$

In summary, this approach uses the reducible polarizability of each subsystem ($\chi^{m/s}$) where the Coulomb potential from the other subsystem is considered part of external potential in Eq. (1), to construct the effective reducible polarizability $\chi_{\text{eff}}^{m/s}$ of each subsystem where such potential is excluded from external potential in Eq. (2). Then we sum up χ_{eff}^m and χ_{eff}^s to obtain total reducible polarizability of interface systems χ^{tot} in Eq. (3), which will be denoted as “ χ_{eff} -sum.”

As we noted above, interlayer wave-function overlap or hybridization effect is not taken into account in the method described above. The hybridization effect can change the eigenvalues and eigenfunctions at the DFT level, which then change the Green’s function (G) and dielectric matrix (in W) in the GW calculations. Therefore, for systems with strong interlayer hybridization, we can add the hybridization effect step by step. We can add corrections from ground-state eigenvalues of interfaces to the χ_{eff} -sum methods, namely “ $\chi_{\text{eff}}^{\text{GSC}}$ -sum” method, which partially take into account the effect of interlayer hybridization on eigenvalues at the DFT level. Furthermore, we can also include interface ground state wave function (“FWF”) and eigenvalues as inputs for Green’s function (G), denoted as the “ $\chi_{\text{eff}}^{\text{FWF}}$ -sum” method. This method is close to GW calculations of an explicit interface except with approximate dielectric matrix by Eq. (3).

From another perspective, if the interlayer hybridization or wave-function overlap is negligible (similar to the condition required above for δn^{tot}) [44,45], the total irreducible polarizability

χ_0^{tot} of the interfaces can be expressed approximately as the sum of each subsystem contribution [15,37,38,44–46],

$$\chi_0^{\text{tot}} = \chi_0^m + \chi_0^s, \quad (4)$$

which we denote as the “ χ_0 -sum” method. To further understand the theoretical connection between different methods, we rewrite Eq. (1) with χ_0 through relation $\chi = \chi_0 + \chi_0 v_C \chi$ as

$$\begin{aligned}\delta n^m &= \chi_0^m(\delta V_{\text{ext}} + v_C \delta n^s + v_C \delta n^m), \\ \delta n^s &= \chi_0^s(\delta V_{\text{ext}} + v_C \delta n^m + v_C \delta n^s).\end{aligned}\quad (5)$$

Here χ_0 as the irreducible polarizability is the density response function to total field δV_{tot} , which includes the applied field and bare Coulomb potential of the total interface system, namely $\delta V_{\text{tot}} \equiv \delta V_{\text{ext}} + v_C \delta n^{\text{tot}}$. Using the above condition $\delta n^{\text{tot}} = \delta n^m + \delta n^s$ for the interface, summation of the two equations of subsystems in Eq. (5) results in $\delta n^{\text{tot}} = \chi_0^{\text{tot}} \delta V_{\text{tot}} = (\chi_0^m + \chi_0^s) \delta V_{\text{tot}}$, which gives Eq. (4). This indicate that the χ_{eff} -sum method and χ_0 -sum method are equivalent under RPA. However, the χ_{eff} -sum method and χ_0 -sum method are not equivalent when the diagonal approximation is applied, i.e., neglecting off-diagonal elements of χ in the former or χ_0 in the latter, as we will discuss in the Sec. II B. Therefore, we primarily used the χ_{eff} -sum method in this paper.

If we further neglect the interlayer Coulomb interaction, this will set $v_C \delta n^{m/s}$ to zero in Eq. (1) and lead to $\chi_{\text{eff}} \rightarrow \chi$. This is at the noninteracting limit between two layers, where

$$\chi^{\text{tot}} = \chi^m + \chi^s, \quad (6)$$

and we name it the “ χ -sum” method. In Sec. IV A, we will compare the quasiparticle energies of interfaces with the above approximated dielectric matrices with explicit interface GW calculations.

B. Diagonal approximation of dielectric screening

For simple metals which may be treated as “jellium,” the nearly translational invariance justifies the dielectric matrix ϵ may be diagonal in reciprocal space [23]. However, semiconductors and insulators that have strong inhomogeneity at interaction length scale require nonzero off-diagonal elements of ϵ [23,47]. The effect from off-diagonal elements of dielectric matrix ϵ is often referred to the “local field effect” [23,47,48].

While the effect of off-diagonal terms in intrinsic dielectric screening has been systematically studied [23,47,48], the off-diagonal terms’ effect from environmental dielectric screening has not been studied in detail. Here we will investigate the off-diagonal effect of environmental dielectric screening through two different approaches, i.e., by applying the diagonal approximation of dielectric matrix ϵ (“ ϵ -diag,” which directly relates to diagonal approximation of χ_0) or inverse dielectric matrix ϵ^{-1} (“ ϵ^{-1} -diag,” which directly relates to diagonal approximation of χ and χ_{eff}).

The ϵ -diag approximation has been used for substrate dielectric screening in past work [15,37,38,46] when applying the χ_0 -sum method, specifically, by removing the in-plane off-diagonal components of substrate dielectric matrices. The

ϵ^{-1} -diag approximation has not been employed before, but it is more convenient in the χ_{eff} -sum approach. Since the off-diagonal elements of χ_0 will contribute to the diagonal elements of χ and ϵ^{-1} through the matrix inverse operation, this is a weaker approximation than ϵ -diag. We compare these two approximations considering specific numerical examples in Sec. IV B.

C. Reciprocal-space linear-interpolation approach

The construction of interface structure models is often complicated by the problem of lattice matching between two subsystems. One of the main objectives of this work is to propose a general approach that can be applied to subsystems with different periodicity and crystal symmetry and that does not require the application of strain to force the lattice matching at the interface.

In general, in order to directly sum the subsystem contributions to obtain the polarizability (and dielectric matrix) of the full interface, one needs an exact correspondence of the $\mathbf{q} + \mathbf{G}$ vectors between the material and the substrate. This requires finding two integer numbers N and $\tilde{N}$ such that the lattice constants L (substrate) and $\tilde{L}$ (material) satisfy the relation $L \cdot N = \tilde{L} \cdot \tilde{N}$. If N and $\tilde{N}$ can be chosen to be reasonably small, calculations can be directly performed for supercells containing N and $\tilde{N}$ repetitions, although this approach often requires the application of a small percentage of strain. However, if the required N or $\tilde{N}$ are large, several methods have been proposed to make this type of calculation practical [15,37,38,44–46]. The central idea is to consider unit cells only and to perform a one-to-one mapping between the reciprocal space $\mathbf{q} + \mathbf{G}$ vectors of the material and substrate [45] [see Fig. 2(a)]. This approach still requires the relation $L \cdot N = \tilde{L} \cdot \tilde{N}$ to be satisfied (possibly by applying a small strain to modify L or $\tilde{L}$) but avoids supercell calculations. We note that even if one applies the diagonal approximation for χ_0 or χ , the diagonal elements still contain both $\mathbf{q}$ and $\mathbf{G}$ vectors, which requires this relation to be satisfied. While this is a clear numerical improvement, a large number of $\mathbf{q}$ vectors in the first Brillouin zone might still be required. Indeed, one needs to sample $\mathbf{q}$ and/or $\tilde{\mathbf{q}}$ point meshes fine enough to ensure that the number of $\mathbf{q}$ points (N_q) satisfy the relation $L \cdot N_q = \tilde{L} \cdot N_{\tilde{q}}$ or equivalently $N_q/N_{\tilde{q}} = N/\tilde{N}$. Accordingly, this approach becomes computationally demanding for large N and/or $\tilde{N}$. A more serious issue is that this mapping scheme is not possible for interfaces with two systems with very different crystal symmetries, e.g., hexagonal and lattice.

In this work, we propose a general method for arbitrarily lattice-mismatched interfaces where it is not possible to map the $\mathbf{q} + \mathbf{G}$ vectors between the two subsystems. This approach applies a linear interpolation of the matrix elements on the substrate grid ($\mathbf{q} + \mathbf{G}$, $\mathbf{q}' + \mathbf{G}'$) to obtain their representation on the material grid ($\tilde{\mathbf{q}} + \tilde{\mathbf{G}}$, $\tilde{\mathbf{q}}' + \tilde{\mathbf{G}}'$), as shown in Figs. 2(b) and 2(c). We note that we need to interpolate $\mathbf{q} + \mathbf{G}$ together between materials and substrates, which can completely remove the symmetry constrain. Interpolation of $\mathbf{q}$ only as done in the past work [49,50] will improve $\mathbf{q}$ -sampling convergence speed but does not solve the periodicity or symmetry mismatch problem at interfaces. As this procedure requires a sampling of the $\mathbf{q}$ vectors over the full

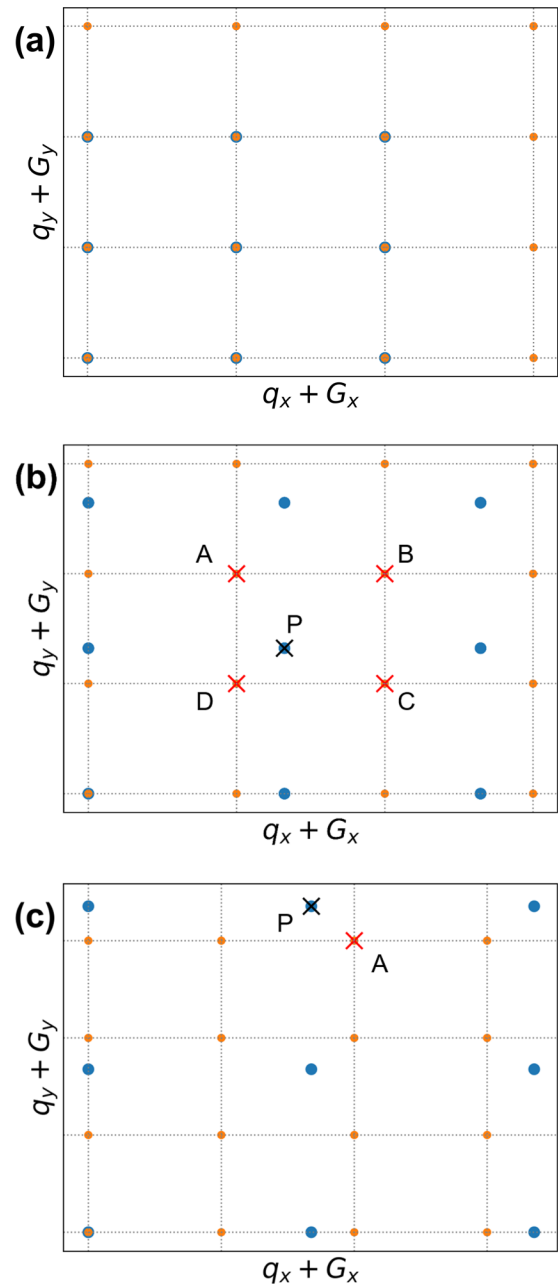


FIG. 2. Schematic diagram of (a) mapping between computed data points of substrate (orange dots) and computed data points of material (blue dots), where the reciprocal space $\mathbf{q} + \mathbf{G}$ grid from the substrate and $\tilde{\mathbf{q}} + \tilde{\mathbf{G}}$ grid from the material are overlapping; (b) bilinear interpolation of the black cross point at P (from $\tilde{\mathbf{q}} + \tilde{\mathbf{G}}$ grid) from the four nearest data points A, B, C, D (red cross) when P is inside the boundary of the $\mathbf{q} + \mathbf{G}$ grid (orange dots); and (c) proximal interpolation with the only nearest one point A when the interpolation point is P at the boundary.

first Brillouin zone (FBZ), whenever necessary, the symmetry operators are used to reconstruct the grid in the FBZ from the grid in the irreducible Brillouin zone (IBZ). Without loss of generality, we choose the same size for vacuum in the z direction for both subsystems; in this way, the same out-of-plane reciprocal lattice G_z components are obtained. In order to simplify the implementation, we neglect the in-plane off-diagonal

elements of the substrate; i.e., we consider $v_C \chi_{\mathbf{G}, \mathbf{G}'}^S(\mathbf{q}) \approx v_C \chi_{\mathbf{G}, \mathbf{G}'}^S(\mathbf{q}) \delta_{G_x, G'_x} \delta_{G_y, G'_y}$. As shown later for specific numerical examples (see Sec. IV C), this approximation works well in practice for mismatched 2D interfaces. For each set of matrix elements at fixed $\{G_z, G'_z\}$, the standard bilinear interpolation technique [51] is used to obtain the corresponding in-plane matrix elements $v_C \chi_{G_z, G'_z}(\tilde{\mathbf{q}}_{x,y} + \tilde{\mathbf{G}}_{x,y})$ in the material subspace, interpolated from $v_C \chi_{G_z, G'_z}(\mathbf{q}_{x,y} + \mathbf{G}_{x,y})$ in the substrate subspace. As shown in Fig. 2(b), the value of the response function $\epsilon^{-1} - 1 = v_C \chi$ at each $\tilde{\mathbf{q}} + \tilde{\mathbf{G}}$ point (denoted by the black cross overlaying the blue dots) is obtained by interpolating the values at the four nearest $\mathbf{q} + \mathbf{G}$ points (denoted by the red cross overlaying the orange dots). We note that the bilinear interpolation method can be applied only if all four nearest neighbours exist within the boundaries of $\mathbf{q} + \mathbf{G}$ space; otherwise, the standard proximal interpolation method is applied, which considers only the nearest point on the grid (most likely at the boundary), as shown in Fig. 2(c). However, the values close to the boundary of $\mathbf{q} + \mathbf{G}$ space are very close to zero as shown in Fig. 3(a). The bilinear interpolation method is fully general regardless of crystal symmetry, which can be applied to arbitrary interfaces.

By applying the interpolation method, we can obtain substrate $v_C \chi$ matrix elements at the material's $\tilde{\mathbf{q}} + \tilde{\mathbf{G}}$ grids, without any artificial strain [15,44,45]. As shown in Fig. 3(a), the orange points are the $v_C \chi_{G_z, G'_z}(\mathbf{q}_{x,y} + \mathbf{G}_{x,y})$ values computed at the substrate momentum space with full BZ, and then we interpolate them to the blue points on the grids of material momentum space $v_C \chi_{G_z, G'_z}(\tilde{\mathbf{q}}_{x,y} + \tilde{\mathbf{G}}_{x,y})$ (only elements in IBZ are shown here). The blue points fall smoothly on the surface of orange points, which show a good interpolation quality. An enlarged picture is also shown in Fig. 3(b). To show the generality of our method, we applied this interpolation method for hBN/phosphorene(BP) interface, where BP has a rectangle lattice, sharply different from the hexagonal lattice hBN has (see Fig. 1 in the Supplemental Material [52]). We show again with our interpolation method that one can obtain the matrix elements of substrates at the material $\tilde{\mathbf{q}} + \tilde{\mathbf{G}}$ grids. Then we can compute the quasiparticle energies of this interface, at two systems' natural lattice constants, with the χ_{eff} -sum method.

III. COMPUTATIONAL DETAILS

A. Computational workflow

The workflow of GW calculations for the interface is structured as follows. We first compute the reducible polarizabilities (χ) for each subsystem separately and then we use them to obtain the effective polarizabilities ($\chi_{\text{eff}}^{m/s}$) using Eq. (2). In the case of lattice mismatch between the two subsystems, the matrix elements of the polarizability χ of the substrates are obtained on the same reciprocal space grid of the material [monolayer (ML) hBN in the practical applications of this work] by using the linear interpolation method described above. Next we sum them to obtain χ^{tot} (i.e., the χ_{eff} -sum method).

Finally, in order to include the screening effect of the substrate on the material, the GW calculations are performed for the standalone hBN ML with the χ^{tot} obtained in the previous step. As we will discuss later, one can achieve further improvement for interfaces with strong hybridization by

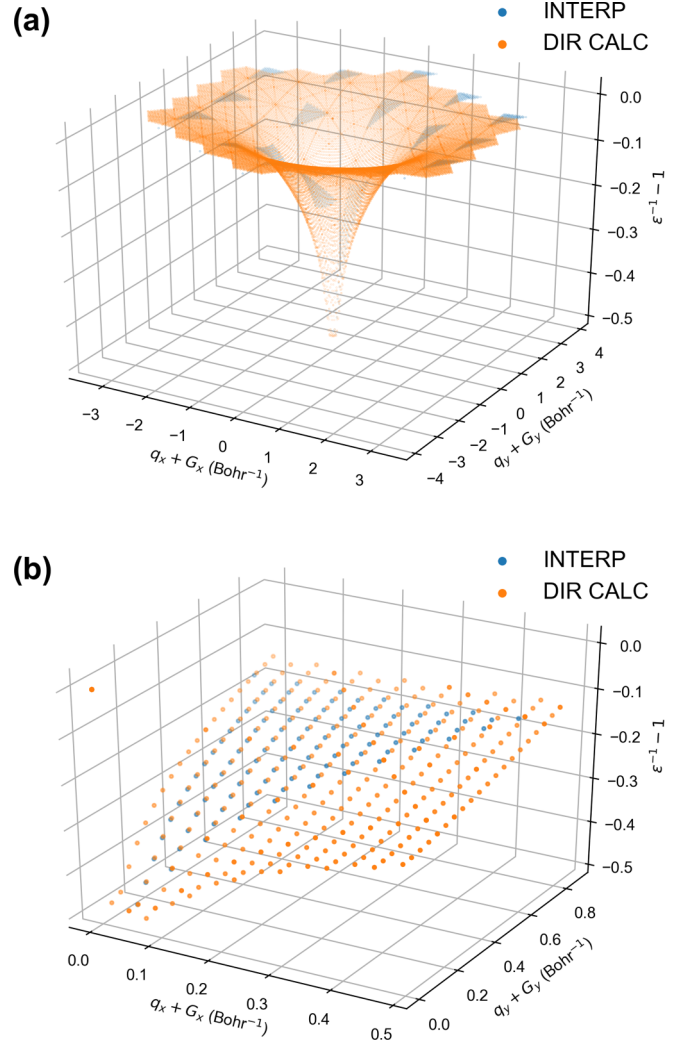


FIG. 3. 3D plot of in-plane diagonal elements of function $\epsilon^{-1} - 1$ of SnS_2 substrate, with $\mathbf{G}$ vector subset $(G_z, G'_z) = (0, 0)$ in (a) full reciprocal space and (b) an enlarged portion of panel (a) that contains the irreducible Brillouin zone of the interpolated points (blue) in hBN $\tilde{\mathbf{q}} + \tilde{\mathbf{G}}$ subspace. The $q_i + G_i$, $i = x, y$ are in-plane reciprocal space Cartesian coordinate in atomic unit (Bohr^{-1}). The orange points are directly computed data points (“DIR CALC”) in SnS_2 substrate $\mathbf{q} + \mathbf{G}$ subspace, while the blue points are interpolated points (“INTERP”) to (hBN) material $\tilde{\mathbf{q}} + \tilde{\mathbf{G}}$ subspace. Note that the orange points used for interpolating blue points in panel (b) are beyond the first Brillouin zone of the substrate $\mathbf{q} + \mathbf{G}$ subspace. The single point at zero is the head element of $\epsilon^{-1} - 1$, which is exactly zero for both material and substrate.

including corrections from ground-state eigenvalues and wave functions of explicit interfaces.

B. Numerical parameters

In this work, we mainly focus on the quasiparticle energies of monolayer hBN/substrate interfaces as prototypical systems (where as substrates we will consider monolayer hBN itself and monolayer SnS_2). Density functional theory (DFT) ground-state calculations based on the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [53] have been performed using the open source plane-wave code

QUANTUM ESPRESSO [54] with optimized norm-conserving Vanderbilt (ONCV) pseudopotentials [55] and a 80-Ry wavefunction cutoff. From structural relaxation, we obtained lattice constants of 2.51 (Å) and 3.70 (Å) for the free-standing ML hBN and SnS₂, respectively.

GW calculations with the Godby-Needs plasmon-pole approximation [56,57] (PPA) were then performed using the YAMBO [58] code. We chose PPA as a showcase for lower computational cost, but we can apply the same $\chi_{\text{eff}}\text{-sum}$ and reciprocal-space interpolation method with full frequency integration as well without technical difficulty, with more computational cost. Importantly, to the best of our knowledge, only PPA models or static coulomb hole plus screened exchange (COHSEX) approximation were used in past calculations for the dielectric screening of substrates [15,36–38,44–46] and the obtained results were reasonably accurate. We used the same plasmon frequency for all calculations $\omega_p = 27.2$ eV and found little variation of the results (i.e., within 20 meV with ω_p from 24.5 to 30 eV).

The distance between the nearest periodic repetitions along the vacuum direction was set to be 20 Å. In order to speed up convergence with respect to vacuum sizes, a 2D Coulomb truncation technique was applied to dielectric matrices and GW self-energies [59]. For bilayer hBN systems, we set the interlayer distance to the bulk value of 3.33(Å) for both of the two different stacking configurations considered here (AA' and AB). The hBN/SnS₂ interlayer distance was set to 3.31 (Å) as obtained from structural relaxation with vdW-corrected functionals [60,61].

For each free-standing ML unit cell, the GW self-energy cutoff is set to 15 Ry. The number of bands is set to 1000 (1500) for hBN (SnS₂) unit cell calculations. The exchange self-energy cutoff is set to 40 Ry. We use a $30 \times 30 \times 1$ ($20 \times 20 \times 1$) $\mathbf{k}$ -point sampling for ML-hBN (ML-SnS₂) unit cell calculations, unless specified.

GW calculations for the full explicit heterointerfaces have also been performed to obtain “exact” reference results to benchmark the different methods for the substrate screening effects (see Sec. IV A). The computational parameters for the full interface are set to keep consistency between supercells and unit cell calculations. Additional computational details and convergence tests can be found in the Supplemental Material [52].

IV. RESULTS AND DISCUSSION

A. Numerical comparison of different methods for substrate screening

After presenting in Sec. II with different approaches to approximate the total dielectric screening of an interface between two weakly interacting subsystems, in this section we discuss their accuracy in practical GW calculations. Results for explicit interfaces will be used as a reference. Specifically, we computed the GW quasiparticle bandgaps of three interfaces: hBN/SnS₂, 2L-AB stacking hBN with two layers' atoms misaligned, and 2L-AA' stacking hBN interface with two layers' atoms aligned (the corresponding atomic structures are shown in Fig. 1). In order to keep the comparison of different methodologies as simple as possible, the calculations in this section are performed with fully commensurate

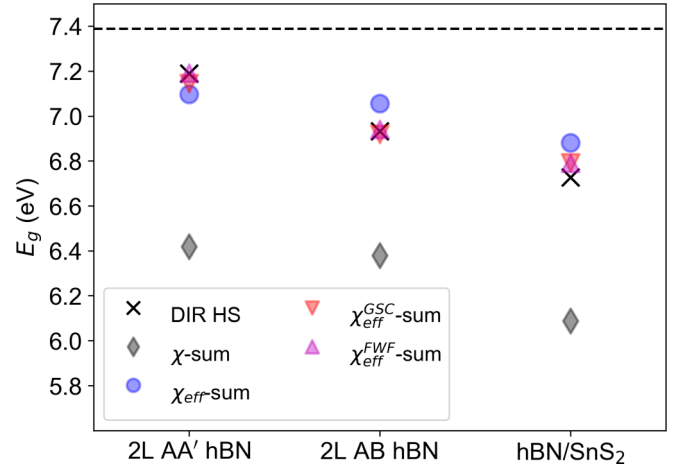


FIG. 4. hBN direct band gap at K from several interfaces with different approximations of substrate screening, compared with explicit interface calculations. The black dashed line is the direct band gap at K of free-standing ML hBN. For each symbol in the figure, “DIR HS” with black cross denotes direct GW calculation of explicit heterostructure; “ $\chi_{\text{eff}}\text{-sum}$ ” with blue circle denotes sum of effective polarizability approach by Eq. (3) with ground-state inputs from free-standing ML hBN; “ $\chi_{\text{eff}}^{\text{GSC}}\text{-sum}$ ” method with red down triangle denotes “ $\chi_{\text{eff}}\text{-sum}$ ” method with additional eigenvalue corrections from ground state interface eigenvalues (“GSC”); “ $\chi_{\text{eff}}^{\text{FWF}}\text{-sum}$ ” with magenta up triangle denotes “ $\chi_{\text{eff}}\text{-sum}$ ” method with both ground-state eigenvalues and wave functions from interfaces; and “ $\chi\text{-sum}$ ” denotes noninteracting “ $\chi\text{-sum}$ ” method by Eq. (6).

interfaces, for both explicit and approximate interface calculations, as shown in Fig. 4.

From the explicit interface results in Fig. 4, we see that the direct band gap of hBN at the hBN/SnS₂ interface (black cross in the third column) is reduced by 0.8 eV compared with the isolated ML hBN (dashed line). This value is about four times of the band-gap reduction for the bilayer hBN with respect to the isolated ML hBN (black crosses in the first and second columns). This is because ML SnS₂ has a much stronger dielectric screening ($\epsilon_{\infty} \approx 17$) and a smaller electronic band gap (≈ 2 eV) compared to ML hBN, which has $\epsilon_{\infty} \approx 5$ and an electronic band gap of ≈ 7 eV. This indicates the positive correlation between electronic band-gap reduction and substrate dielectric screening, similar to previous discussions [62–64].

Second, we find that the effective polarizability approach results ($\chi_{\text{eff}}\text{-sum}$ method, blue circle) are consistently in good agreement with the ones from explicit interface GW calculations (direct, black cross), i.e., within 0.2 eV. We improve the agreement by 50 meV with additional corrections from ground-state eigenvalues of interfaces ($\chi_{\text{eff}}^{\text{GSC}}\text{-sum}$ method, red triangle), which partially take into account the effect of interlayer couplings on eigenvalues at the DFT level. Moreover, by using interface ground-state wave functions and eigenvalues (FWF) as inputs for Green’s function calculations, the results of the effective polarizability approximation ($\chi_{\text{eff}}^{\text{FWF}}\text{-sum}$ method, green square) are further improved, i.e., with only 10 meV difference from the explicit interface GW calculations. While a similar approach was used in Ref. [44], the $\chi_{\text{eff}}^{\text{FWF}}\text{-sum}$ method has a computational cost similar to that

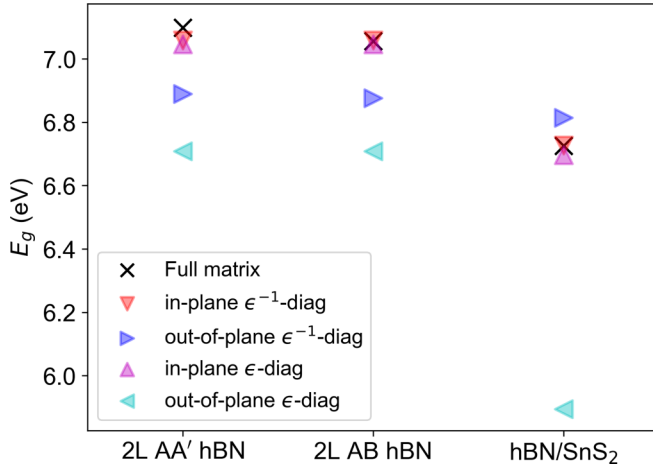


FIG. 5. GW results for hBN direct band gap at K using χ_{eff} -sum or equivalently χ_0 -sum method to examine the effect of diagonal approximations. “Full matrix” in black cross denotes full dielectric matrices without any diagonal approximation as reference; “in-plane ϵ^{-1} -diag” in red downward-pointing triangle denotes diagonal approximation to in-plane elements of ϵ^{-1} ; “out-of-plane ϵ^{-1} -diag” in dark blue right-pointing triangle denotes diagonal approximation to out-of-plane elements for ϵ^{-1} ; “in-plane ϵ -diag” in magenta upward-pointing triangle denotes diagonal approximation to in-plane elements of ϵ ; and “out-of-plane ϵ -diag” in light blue left-pointing triangle denotes diagonal approximation to out-of-plane elements of ϵ .

of the full interface GW calculation (although the evaluation of the dielectric matrix is more efficient) and is much more demanding than the other methods in Fig. 4 and Table I. Therefore, χ_{eff} -sum and $\chi_{\text{eff}}^{\text{GSC}}$ -sum provide the best compromise between accuracy and computational cost. We note that for explicit hBN/SnS₂ interface, we had to apply 1.5% strain to obtain commensurate supercells, which may explain why this interface has slightly larger difference between χ_{eff} -sum and explicit calculation than bilayer hBN.

In sharp contrast to the methods discussed above, the noninteracting interlayer method based on Eq. (6) (χ -sum method, black diamond) gives results far from the explicit interface reference (e.g., with an error of about 0.6 eV). This indicates that the interlayer Coulomb interaction plays a dominant role in the electronic band-gap reduction by substrate screening.

B. Diagonal approximation of substrate dielectric screening

In this section, we will compare different diagonal approximations for the screening considering different numerical examples. With “in-plane ϵ^{-1} -diag,” we will denote an approximation that discards the in-plane off-diagonal elements of reducible polarizability χ in reciprocal space, i.e., $\chi_{\mathbf{GG}'}(\mathbf{q}, \omega) \delta_{G_x, G'_x} \delta_{G_y, G'_y}$. Similarly, “out-of-plane ϵ^{-1} -diag” will denote an approach that does not include the out-of-plane off-diagonal elements of polarizability in reciprocal space, i.e., $\chi_{\mathbf{GG}'}(\mathbf{q}, \omega) \delta_{G_z, G'_z}$. Analogous definitions will be used for ϵ -diag.

The GW quasiparticle gaps with different diagonal approximations for the hBN bilayer in two different conformations (AA'/AB) and the hBN/SnS₂ interface are shown in Fig. 5.

We find that for both the ϵ^{-1} and ϵ -diagonal approximations, neglecting out-of-plane off-diagonal elements of the substrate (“out-of-plane ϵ^{-1} -diag” and “out-of-plane ϵ -diag,” denoted by dark blue right-pointing triangle and light blue left-pointing triangle, respectively) causes a large discrepancy of the bandgaps (i.e., from 0.2 to 0.8 eV) with respect to the “exact” result obtained from the full screening matrix (“full matrix,” black cross). In contrast, the results obtained by neglecting in-plane off diagonal elements (in-plane ϵ^{-1} /in-plane ϵ -diag, red downward-pointing triangle and magenta upward-pointing triangle) are similar to those with the full screening matrix with deviations within 50 meV. This means the inhomogeneity effect of out-of-plane substrate screening on quasiparticle energies is much stronger than the one of in-plane substrate screening, because the out-of-plane direction is along the nonperiodic (vacuum) direction with dramatically inhomogeneous charge distribution, compared to the in-plane periodic direction.

Besides the overall difference of diagonal approximation along different directions, we also distinguish the difference between ϵ^{-1} -diag and ϵ -diag approaches in each case. (1) Along the in-plane direction, the difference between different approaches is negligible, i.e., less than 10 meV. (2) Along the out-of-plane direction, the out-of-plane ϵ^{-1} -diag results (dark blue rightward-pointing triangle) are much closer to the full dielectric matrix results (black cross) than the out-of-plane ϵ -diag results (light blue left triangle) in Fig. 5. This is consistent with our earlier speculation that the ϵ^{-1} -diag may be a better (weaker) approximation, because the off-diagonal elements of irreducible polarizability χ_0 contribute to χ or ϵ^{-1} during its matrix inversion, which is completely missing in the ϵ -diag approximation.

Moreover, the in-plane inhomogeneity is relatively larger when there is stronger interlayer coupling with atoms aligned perfectly for chemical bonding. For example, the in-plane inhomogeneity of bilayer hBN with atoms aligned [e.g., 2L AA' hBN in Fig. 1(b); both in-plane ϵ^{-1} -diag (red downward-pointing triangle) and in-plane ϵ -diag (magenta upward-pointing triangle) results have 40-meV difference from the “full matrix” results in the first column of Fig. 5] is larger than the interfaces with atoms misaligned [e.g., 2L AB hBN and hBN/SnS₂ heterostructure in Figs. 1(a) and 1(c); both in-plane ϵ^{-1} -diag and in-plane ϵ -diag results have no difference from “full matrix” results in the second and third columns of Fig. 5].

C. Lattice mismatched hBN/SnS₂ interface

In order to benchmark the reciprocal-space linear-interpolation method introduced in Sec. II C and Table I, we consider the hBN/SnS₂ interface. A strain of 1.5% was applied to SnS₂ to match the hBN lattice constant with a 2:3 ratio in each direction of the plane (namely $2L^{\text{SnS}_2} = 3L^{\text{hBN}}$). By using a commensurate $\mathbf{q}$ -point sampling for the two subsystems with a 2:3 ratio, a mapping of the $\mathbf{q} + \mathbf{G}$ vectors is possible and traditional methods for the substrate effect can be applied to produce a reference results for our new interpolation method (which, instead, will be used with an incommensurate $\mathbf{q}$ -point sampling). We computed the GW band edges near the high-symmetry point K of hBN on the

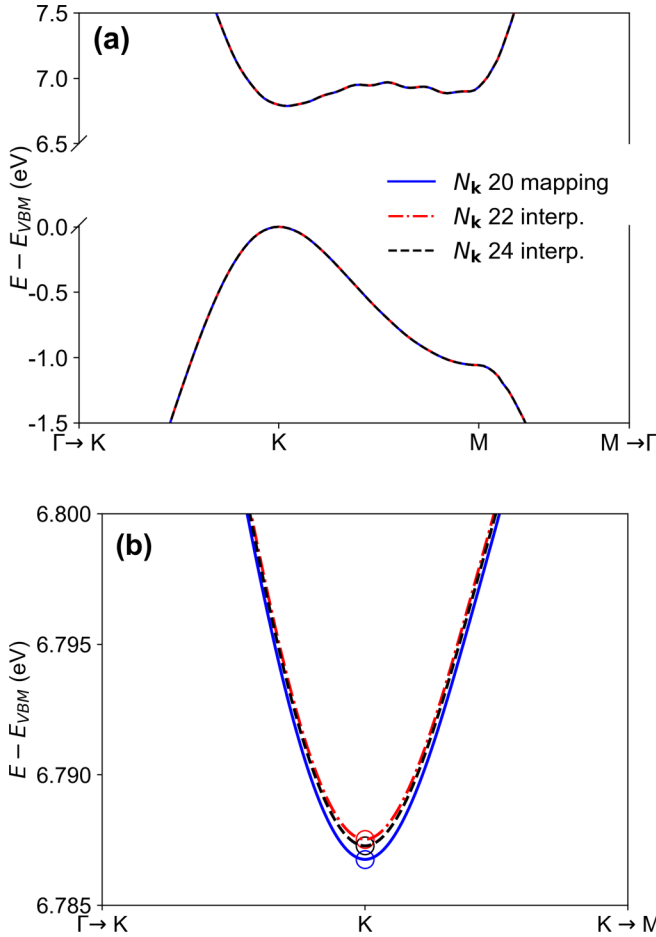


FIG. 6. GW band structure of hBN at hBN/stretched SnS_2 interface, referenced to valence band maximum (VBM) by using “ $\chi_{\text{eff}}\text{-sum}$ ” method. The blue solid line is computed with commensurate $\mathbf{q}$ -point sampling with the reciprocal space mapping approach, while the red and black dashed line results are computed with incommensurate $\mathbf{q}$ -point sampling with the reciprocal-space linear-interpolation approach. Panel (a) shows both valence and conduction band edges; panel (b) shows only the conduction band edge close to K.

SnS_2 substrate with the $\chi_{\text{eff}}\text{-sum}$ method at different $\mathbf{k}$ -point samplings, as shown in Fig. 6. The mesh for $\mathbf{q}$ -point sampling was chosen to be identical to the $\mathbf{k}$ -point sampling. Specifically, the reference calculations were performed with the hBN unit cell calculation with $20 \times 20 \times 1$ and $30 \times 30 \times 1$ $\mathbf{k}$ -point sampling for the units cells of SnS_2 and hBN, respectively (this choice satisfies the 2:3 ratio for each in-plane direction). The reference result obtained from the $\mathbf{q} + \mathbf{G}$ mapping is shown in Fig. 6 (blue curve labeled by “ $N_{\mathbf{k}}$ 20 mapping”). To apply our interpolation technique, it is not necessary to use commensurate grids and we compare instead the results for two different choices of the $\mathbf{q}$ -point sampling for SnS_2 . Specifically, in Fig. 6 we show the results for the $22 \times 22 \times 1$ (red dashed line, “ $N_{\mathbf{k}}$ 22 interp.”) and $24 \times 24 \times 1$ (black dashed line, “ $N_{\mathbf{k}}$ 24 interp.”) $\mathbf{q}$ -point grids, which do not allow for a mapping of the reciprocal space vectors and would be impossible to treat without our interpolation method. The results in Fig. 6(a) show that the GW band structure with interpolation (red and black dashed lines) is nearly identical to the

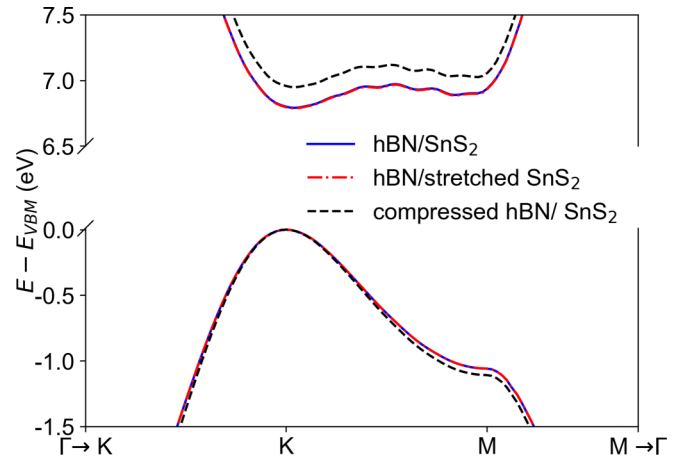


FIG. 7. GW band structures of hBN with stretched SnS_2 substrate (“hBN/stretched SnS_2 ,” red dash-dotted line), hBN with SnS_2 substrate with no strain (“hBN/ SnS_2 ,” blue solid line), and compressed hBN with SnS_2 substrate (“compressed hBN/ SnS_2 ,” black dashed line), respectively.

one based on the mapping (blue solid line), with differences smaller than 1 meV [as can be seen by enlarging the conduction band edge in Fig. 6(b)]. This comparison demonstrates the excellent numerical accuracy of our linear interpolation method, which could have also been expected from the high quality of the interpolation in Fig. 3.

Finally, we use our interpolation method to better understand the effect of the strain on quasiparticle energies. In Fig. 7, the blue curve corresponds to the the hBN band structure on the SnS_2 substrate without strain for either system as obtained from the interpolation scheme described above. These results are compared with those obtained for the interface by applying strain either to compress the hBN lattice parameter or to stretch SnS_2 . We found that even a 1.5% compressive strain for hBN (black dashed line), the conduction band edge changes by 0.2 eV. Since we are focusing on the band structure of hBN states, the application of the strain to the SnS_2 substrate leads to negligible changes (red dash-dotted line). This result highlights the high sensitivity of quasiparticle band structures to strain.

We note that for a proof of principle and benchmark purpose, we chose systems with similar crystal symmetry, i.e., hexagonal lattice, in this work. However, our interpolation method can be applied to general interfaces with very different crystal symmetry, e.g., interface between hexagonal and rectangle lattices, as the example of hBN/phosphorene interface shown in the Supplemental Materials Fig. 1 [52]. This is not possible by using the previous $\mathbf{q} + \mathbf{G}$ mapping approach. Our reciprocal-space linear-interpolation method makes possible the GW calculations of interfaces composed by two materials with very different lattice parameters and symmetry, at the cost of primitive cell calculations only.

V. CONCLUSION

In this work, we theoretically and numerically examined the existing methods to approximate substrate dielectric screening effect on quasiparticle energies, through hBN heterostructures as prototypical examples. We clarified the

theoretical equivalence between the sum of effective reducible polarizability approach ($\chi_{\text{eff}}\text{-sum}$) and sum of irreducible polarizability of interface systems ($\chi_0\text{-sum}$) at the RPA level. We numerically compared the GW calculations of 2D interfaces with several approximations and found excellent agreements between $\chi_{\text{eff}}\text{-sum}$ and the explicit interface calculations. Further improvement can be achieved by including the ground-state corrections of eigenvalues (and wave functions) from explicit interfaces. We further evaluated the importance of nondiagonal elements of ϵ and ϵ^{-1} from substrates on quasiparticle energies of 2D interface. Most important, we developed an accurate reciprocal-space linear-interpolation technique for arbitrarily lattice-mismatched interfaces, which can be used to compute the interface polarizability for GW quasiparticle energies without any artificial strain, at the cost of only primitive cell calculations.

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