Bidimensional materials are, as their name suggests, ideally viewed as having no thickness. The advent of multilayer stacks of 2D materials and combinations of different materials in vertical van der Waals heterostructures highlights however that these materials have a finite thickness. In this article, we show how volume properties of stacked 2D layers can be calculated from boundary conditions and conversely. We introduce the layer, surrounded by vacuum, as a kind of transfer matrix with intrinsic parameters. This provides a link between continuous and discrete media, and a connection with the interface reflection and transmission coefficients calculated from microscopic models. We show how to model hybrid systems and identify in the zero-thickness limit the intrinsic parameters of the current sheet that represents the 2D material, namely the in-plane surface susceptibility $\chi^s$ and the out-of-plane parameter $\xi^s$ that corresponds to a displacement susceptibility. By considering anisotropic layers under the assumption that TE and TM modes are not coupled, we provide a unified vision of the different models used in optical characterization of 2D materials, including ellipsometry. We build on this to model 3D structures layer per layer and identify their effective permittivity and propagation constants. We show that our model fits existing ellipsometric data with the same reliability as the existing interface models, but with the advantage that multilayer and monolayer systems are described in a same way.

I. INTRODUCTION

Following the important impact of 2D materials in different domains of applications such as electronics, photonics, biomedical engineering, printing technology and aerospace [1], multilayer systems are progressively developing. They include vertical van der Waals heterostructures [2,3] and could be used to increase the total layer response [4,7]. study the interaction between layers [4], protect against electromagnetic microwave pollution [8], or to devise new materials combining their properties by stacking them together.

Optical properties of single- and multilayer systems can be used to probe the number of layers [9,10] or investigate the evolution of the refractive and conductive properties with the number of layers [4,7,11]. In waveguiding optics, 2D materials or multilayer systems are useful for functionalization [12], modulation [13,14] or to enhance the nonlinear properties [15,18].

With their atomic thickness, 2D materials can be described either by surface currents at an interface or as ultrathin films of continuous materials [1]. These two descriptions have been used in parallel, raising questions about which model is more physically sound and why they differ on their predictions [19].

A first attempt to merge the two models in a single picture was provided by Majérus et al. in [20] within an Anisotropic Interface Model (AIM), where the importance of the anisotropy of the thin-film model and of the out-of-plane component of the surface susceptibility was pointed out. In a very nice experimental work [21], Xu and co-workers highlighted the importance of the out-of-plane component, and Dell’Anna showed how to model this out-of-plane response from a microscopic point of view in a Microscopic Reaction Model (MRM) [22]. Although the work in [20] and [21] present similarities, they do not provide the same analytical expressions for the reflection coefficient, and the first of these models is not purely conservative for a lossless 2D material immersed in a lossless medium. More over, while these two works describe the impact of a 2D material at the interface between two continuous bulk media, they do not provide a clear way to model multilayer systems. Tab. I summarizes the main characteristics of the different models that are depicted on Fig. 1. It shows that existing models, such as the AIM and MRM describe well the boundary conditions at an interface, but not the multilayer systems; while the Isotropic Transfer Matrix (ITM) model can describe multilayer systems but without anisotropy. The Anisotropic Layer Model (ALM) that will be presented below combines all the advantages.

The first aim of this work is to provide practical expressions to relate microscopic boundary conditions to the properties of a single-layer or multilayer system. These expressions will be cast on the form of a transfer matrix that is very useful in practical applications such as ellipsometry. In doing this, we will also show how the two models of Majérus [20] and Dell’Anna [22] differ, and how they can be adapted to provide a unified description of a single 2D layer or a multilayer system of stacked materials. To reach this goal we will have to identify intrinsic parameters of a layer, i.e. parameters that do not depend on the surrounding medium. We will base our approach on an ALM.

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TABLE I: Main characteristics of the existing models. AIM [20]: Anisotropic Interface Model; MRM: Microscopic Reaction Model [21, 22]; ITM: Isotropic Transfer Matrix [23]; ALM: Anisotropic Layer Model (Sec. III). Comparison of the main models with respect to their ability to describe: (i) an interface with zero-thickness (boundary conditions); (ii) a single layer with a non-zero thickness; (iii) multilayer stacks; (iv) the in-plane – out-of-plane anisotropy of the material; and finally, to provide expression that ensure flux conservation in lossless systems.

| Model | Interface layer | multilayer | anisotropy | conservative |
|-------|----------------|------------|------------|--------------|
| AIM   | +              | -          | +          | -            |
| MRM   | +              | -          | -          | +            |
| ITM   | -              | +          | +          | -            |
| ALM   | +              | +          | +          | +            |

Our approach will be analytical in the first place. It will be compared with existing experimental results. We will show that different models can fit the experimental curves and we will select the most appropriate model by applying a very simple criterion: the description of a N-layer material should be the same as the description of a (N-1)-layer material plus a single layer, i.e. there is not reason to handle the one-layer problem differently from the N-layer one.

Our analysis will be performed assuming that there is no change of the intrinsic parameters with the number of layers. We will on purpose ignore inter-layer effects [4] that could lead to new physics related to the stacking orientation, so that experimental deviations from the optical model of propagation can be attributed to such interactions.

A second aim of the present work is to provide expressions of the interface conditions that are compatible with the approaches of Xu and Majérus. We will build our model step by step, combining transfer-matrices, with the boundary conditions obtained from microscopic modelling.

In Section II we introduce the boundary conditions at an interface that contains an anisotropic 2D material modeled with surface currents. Next, we introduce the concept of layer in Sec. III showing how the volume and the layer properties are related in a continuous and in a discrete medium. In Sec. IV and VI we relate the propagation constant and Fresnel coefficients to the boundary conditions, allowing to calculate volume properties from the boundary conditions. This is applied in Sec. VII to a hybrid material for which we calculate effective parameters, which is very instructive to identify intrinsic parameters in Sec. VIII. Our analytical layer model is then compared to the existing models used for fitting experimental data in Sec. IX. These models are of two different types: interface and layer ones. We show that both are compatible with the experiments, although the layer model appears more general. After that, we present and compare different ways to model a multilayer system in Sec. X, where we also discuss their differences. Finally, we summarize our main findings and conclude.

FIG. 1: From left to right: interface model, continuous layer model, discrete layer model. Surrounding materials are labeled a and b. The interface (left) is described with a surface susceptibility $\chi_s$ (red line). The layer can be described either as a continuous medium (center) or as a discrete medium (right). The continuous layer (center) is described by a permittivity $\varepsilon_m$ in a thin film. The discrete layer (right) is modeled using a current sheet with surface susceptibility $\chi_s$ (red line) surrounded with vacuum ($\varepsilon_0$) to take into account the thickness $d$ of the 2D material.

II. CONTINUITY CONDITIONS BETWEEN ANISOTROPIC MEDIA

Continuity conditions and translational invariance allow to calculate the evolution of an electromagnetic wave at a planar interface between homogeneous media. In particular, without surface currents, tangential components of the electric and magnetic fields are continuous, as well as normal components of the induction and displacement fields. This is not valid anymore in the presence of an interface 2D layer. As shown in [17, 20, 24] using distributions, and in [21, 22] using a microscopic approach involving reactions fields, a surface polarization field orthogonal to the interface appears in this case: $\mathcal{P}_\perp$.

There is an important difference between the microscopic approach of Dell’Anna [22], and that of Sipe [17, 24], in that the first one provides an explicit expression for the discontinuity at the interface, while the second one does not specify how to evaluate the electric field at the interface $\mathcal{E}_i$ to take into account to calculate the surface polarization field from $\mathcal{P}_\perp = \varepsilon_0 \mathcal{P}_\perp \mathcal{E}_s$ (with $\mathcal{P}_\perp$ the surface susceptibility tensor). Possible choices include the electric field in medium a (see Fig. 1), the one in medium b or an average of the two.

In [20], using the approach of [25], the average of the electric field in the surrounding media was used, while in the microscopic approach of [22] the surface field is equal to the transmitted field.

We will see in Sec. III that there is another possibility, that will enable to describe a material made of stacked layers exactly as a continuous material. This will introduce the intrinsic surface quantity $\xi_s$, the displacement susceptibility.
A. Anisotropic boundary conditions

The geometry that we consider here is depicted on Fig. 1 (right panel). As detailed in [20], and summarized in appendix, anisotropic boundary conditions at a 2-D interface between media \(a\) and \(b\) on which a monochromatic plane wave is incident write

\[
1 - \frac{r}{t} = \frac{1}{t} \left( 1 - \frac{1}{r} \right), \quad \psi_{ab} \left( \frac{1}{t} \right), \quad (1)
\]

\[
\alpha_{ab} + \frac{r}{t} = \frac{1}{t} \left( 1 - \frac{1}{r} \right), \quad \psi_{ab} \left( \frac{1}{t} \right), \quad (2)
\]

where the coefficients of transmission \(t\) and reflection \(r\) and the parameter \(\alpha_{ab}\) are defined differently for TE (\(ab\)) waves [see (A.8) and (A.11)]. To avoid coupling between TE and TM waves, we assume that the permittivity tensor is orthogonal when written in the axes \(\chi_z\). This kind of matrix of an interface as shown in Fig. 1. This layer includes interfacial when written in the axes \(\chi_z\) as it is intrinsic to a layer.

B. Boundary conditions and transfer matrix

A transfer matrix links the forward (\(F\)) and backward (\(B\)) components of a plane wave, so that the components in the input medium \(a\) are obtained by multiplying the matrix of the system \(a_{ab}\) with the components at the output medium \(b\).

\[
\begin{pmatrix}
F_a \\
B_a
\end{pmatrix} = \begin{pmatrix}
1 & -r' \\
\frac{F}{r} & \frac{1}{r}
\end{pmatrix} \begin{pmatrix}
F_b \\
B_b
\end{pmatrix},
\]

where \(r\) and \(t\) are the transmission and reflection coefficients appearing in (1) and (2), and their primed versions are calculated by swapping indices \(a\) and \(b\).

Transfer matrix coefficients are easy to calculate if we rewrite (1) and (2) as

\[
\begin{align*}
\frac{1}{t} &= 1 + \frac{\alpha_{ab}}{2} - \frac{1}{2} \varphi_{ab} - \frac{1}{2} \psi_{ab}, \\
\frac{r}{t} &= 1 - \frac{\alpha_{ab}}{2} - \frac{1}{2} \varphi_{ab} + \frac{1}{2} \psi_{ab}. 
\end{align*}
\]

This form is also very interesting to compare analytical predictions of different models, as they are usually provided in terms of \(r\) and \(t\) coefficients. In particular, when the 2D material is immersed in a single material \(a_{ab} = a_{aa} = 1\),

\[
\begin{align*}
t &= t' = 1 + \frac{\varphi_{aa}}{2} + \frac{1}{2} \psi_{aa}, \\
r &= r' = -\frac{1}{2} \varphi_{aa} + \frac{1}{2} \psi_{aa}. 
\end{align*}
\]

These expressions can directly be compared to those of the microscopic model [21]. Although these equations differ, they are the same to the first order in \(\varphi_{ab}\) and \(\psi_{ab}\) provided that \(\tilde{\mathcal{T}} = \epsilon_0 \tilde{E}_b\) (see [26 Sec. VII]). In [20], a coefficient \(c_{ab}\) was used to calculate the effective surface field. It was defined as \(2e_{ab}^{-1} = (e_{ab}^2 / \epsilon_0) - 1 + (e_{ab}^2 / \epsilon_0)^{-1}\). The microscopic model [21] corresponds to the choice \(e_{ab} = e_{ab}^b / \epsilon_0\). In our search for intrinsic parameters, we show in the next section that another choice is necessary.

C. Displacement susceptibility

In Sec. [11] we will describe materials in terms of intrinsic properties of the layers, i.e. parameters that do not depend on the surrounding layers. Although we consider an interface with surface currents, we expect the phase shift induced by the currents to be small, as discussed in [23] and it can be neglected to calculate the field in the layer. Under this approximation, the tangential part of the electric field \(\tilde{E}_g\), and the normal component of the displacement field \(D_z\), are continuous across the interface. This leads to describe the surface polarization field as

\[
\tilde{\mathcal{P}}_g = e_0 \tilde{E}_b, \quad (8)
\]

\[
\tilde{\mathcal{P}}_z = \xi_z \tilde{D}_z, \quad (9)
\]

The \(\xi_z\) parameter, the displacement susceptibility, is dimensionless and can be linked to the expressions of \(\chi_z\) published in [20][22][27]. It appears in first-principle modelling of the out-of-plane response of 2D materials [19][20]. We will see in Sec. [11] that \(\xi_z\) is more suited to describe multilayer systems than \(\chi_z\), as it is intrinsic to a layer.

III. THE ANISOTROPIC LAYER MODEL

In what precedes, we have obtained the matrix for an interface of zero thickness. This kind of matrix of an interface is difficult to use in order to describe a bulk material made of stacked 2D layers, as it would result in a material with a total zero thickness. We therefore define an anisotropic layer with finite thickness but still strongly related to the interface model, that will prove an interesting alternative to the description in terms of interfaces and propagation matrices.

A. Definition

We define our system as a layer with a given thickness \(d\) and an effective permittivity tensor \(\tilde{\mathcal{E}}\) surrounded by vacuum. In what follows, we assume that this tensor is diagonal when written in the axes \(Oxyz\), where \(z\) is orthogonal to the interface as shown in Fig. 1. This layer includes interface matrices to model a homogeneous anisotropic material surrounded by vacuum. The matrix of this layer is given by

\[
\begin{pmatrix}
\mathcal{L}_m(d) \\
\mathcal{P}_m(d), \mathcal{M}_m(d), \mathcal{J}_m
\end{pmatrix} = \begin{pmatrix}
\frac{1+a}{2} - \frac{a-1}{2} \\
\frac{1+a}{2} + \frac{a-1}{2}
\end{pmatrix} \begin{pmatrix}
e^{-i\Phi} & 0 \\
e^{i\Phi}
\end{pmatrix} \begin{pmatrix}
\frac{1+a}{2a} & \frac{a-1}{2a} \\
\frac{a-1}{2a} & \frac{1+a}{2a}
\end{pmatrix}
\]

\[
\begin{pmatrix}
\frac{1}{2} - \frac{1}{2} \\
\frac{1}{2} + \frac{1}{2}
\end{pmatrix} \begin{pmatrix}
\cos \Phi \frac{1+a}{2} + \frac{a-1}{2} \\
\sin \Phi \frac{1+a}{2} + \frac{a-1}{2}
\end{pmatrix} \frac{e^{-i\Phi}}{\sin \Phi}
\]

\[
\begin{pmatrix}
\frac{1+a}{2} & i \frac{a-1}{2} \\
-i \frac{a-1}{2} & \frac{1+a}{2}
\end{pmatrix}
\]

\[
 \frac{e^{-i\Phi}}{\sin \Phi}
\]

\[
\begin{pmatrix}
\cos \Phi & 0 \\
-i \frac{a-1}{2a} \sin \Phi & \sin \Phi \frac{1+a}{2} + \frac{a-1}{2}
\end{pmatrix}
\]

where \(\mathcal{I}_m\) is the usual interface matrix separating homogenous media \(m\) and \(n\), \(\mathcal{P}_m(d)\) is a propagation matrix in medium \(m\) over a distance \(d\), and we have defined \(\alpha = a_{vm} = 1 / a_{mv}\) and written \(\Phi = k_m^m d\) the phase shift induced
by the propagation. Detailed expressions for $\alpha_m$ and $k_z^m$ are given in appendix.

We will show that the matrix of a layer of thickness $d$, seen as a continuous medium is equivalent to the matrix of a zero-thickness current sheet surrounded by two vacuum layers of thickness $d/2$. This explains the dual representation of the ALM on Fig. 1.

It is important to notice that usual thin film models differ from our definition a layer as they consider a transfer matrix that we obtain has the same form as the initial layer matrices, but with a total thickness $d_{tot} = d_1 + d_2$. Layers with different permittivity tensors can also be combined. At the boundary, interface matrices of the two layers combine as

$$\mathbf{I}_{mn}\mathbf{I}_{nm} = \mathbf{I}_{mn},$$

(14)

C. Building a volume material layer per layer

To describe propagation in a bulk material, we could divide it in layers and stack them step by step as shown on figure. Considering an arbitrary number $N$ of layers in the volume, we can write the volume matrix $\mathbf{V}_m$ depending on the thickness $L = Nd$, as

$$\mathbf{V}_m(Nd) = [\mathbf{L}_m(d)]^N = [\mathbf{I}_{vm}\mathbf{P}_m(d)\mathbf{I}_{mv}]^N \mathbf{L}_m(Nd).$$

The impact of layer $N + 1$ is therefore given by

$$\frac{\delta \mathbf{V}_m}{\delta L} = \frac{\mathbf{V}_m(L + d) - \mathbf{V}_m(L)}{d} = \frac{\mathbf{L}_m(d) - 1}{d} \mathbf{V}_m(L).$$

(17)

If the medium is continuous, the number of layers can be chosen high enough to let $d \to 0$, so that, using (11) and $\Phi = k_z^m d$, (17) becomes

$$\frac{d\mathbf{V}_m}{dL} = K_m \mathbf{V}_m,$$

(18)

$$K_m = \lim_{d \to 0} \frac{\mathbf{L}_m(d)-1}{d} = ik_z^m \mathbf{I}_{vm} \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix} \mathbf{I}_{mv},$$

(20)

as $\lim_{d \to 0} (\exp(\pm ik_z^m d) - 1)/d = \pm ik_z^m$. The matrix $K_m$ contains all the information that is needed to build the volume layer. As it is obtained in the limit of small $d$, it can be calculated from (12) or from its first order Taylor expansion in $d$ or $\Phi$, showing that this first order expansion contains all the physics of the system. In this limit,

$$\mathbf{L}_m(d) = I + dK_m.$$  

(21)

This last expression is interesting as it can be computed either from the continuous medium approach, or from the boundary conditions. Indeed, it was shown in [20] that to the first order in $d$ the thin film model and the interface model coincide and then

$$\mathbf{L}_m = \mathbf{I}_{vm}\mathbf{P}_m(d)\mathbf{I}_{mv} = \mathbf{P}_v(d/2)\mathbf{I}_{vv}\mathbf{P}_v(d/2),$$

(22)

where the interface matrix with surface currents $\mathbf{I}_{vv}$ is built from the reflection and transmission coefficients in [4] and [5], in the case where $a$ and $b$ correspond to vacuum. According to [20], the equivalence of the two matrices occurs when

$$\frac{\chi_z^p}{d} = \frac{\mathcal{E}_0^p}{\xi_0^p} - 1,$$

(23)

$$\frac{\chi_z^s}{d} = \frac{\mathcal{E}_0^s}{\xi_0^s} - 1,$$

(24)

$$\frac{\chi_z^s}{d} = \frac{\mathcal{E}_0^s - \mathcal{E}_0^p}{\xi_0^s},$$

(25)

with $\chi_z^s = \mathcal{P}_v / (\mathcal{E}_0^s \mathcal{E}_0^p) \mathcal{P}_v = D_0^s = \mathcal{E}_0^s$, as in vacuum, $D_0^s = \mathcal{E}_0^s \mathcal{E}_0^p$. We can therefore build $K_m$ in two different ways that provide a direct link between the boundary conditions at the interface and the anisotropic thin film model. Our approach differs from [20], in that surrounding materials used
in the calculation of $\mathcal{S}$ are vacuum, whatever the actual surrounding material. Indeed, one goal in [20] was to replace a surface current by a thin film of arbitrary thickness, for example in numerical simulations. Here, we look for intrinsic parameters and we focus only on the exact geometry, where $d$ is the effective thickness of one layer observed, for example, through high-resolution transmission electron microscopy [4].

### D. Volume parameters from a single layer

Our main goal in this article is to build a multilayer matrix from a single-layer one. Here, we will apply the approach of the previous section in the approximation of the continuous volume, where $d$ can be chosen arbitrarily small. Eq. (18) is solved using matrix exponentials. As shown in [26, Sec. IV], we get

$$V_m(L) = e^{K_m L} V_m(0) = \mathcal{J}_{pm} \left( e^{-i k^n_m L} 0 \ 0 e^{i k^n_m L} \right) \mathcal{J}_{mv}. \tag{26}$$

Although this result is not surprising, as it corresponds to a layer matrix $\mathcal{L}_m(L)$, it is very instructive to demonstrate that $K_m$ corresponding to a microscopic thickness $d \to 0$, is sufficient to describe an homogeneous material of macroscopic thickness $L$. It allows to retrieve the propagation matrix and the interface matrices. This will be used in Sec. VII to build the propagation matrix of an hybrid material and identify its effective permittivity tensor.

### E. Discrete medium

In Sec. III C, the integration process was rigorous as we could evaluate (16) in the limit $d \to 0$. The imaginary exponential appearing in [26] is a direct consequence of the convergence of

$$\lim_{N \to \infty} (1 + i k L / N)^N = \exp(i k L). \tag{27}$$

When modelling a 2D material the minimal size of the layer is fixed by the effective thickness of one layer $d_m$. We cannot consider the limit $d \to 0$ anymore. This introduces a discretization error that we will evaluate. To this end, we approximate $K_m$ by taking the first order expansion of $\mathcal{L}_m(d)$ and using

$$K_m = \frac{\mathcal{L}_m(d_m) - I}{d_m}. \tag{28}$$

As the number of layers is fixed by the thickness of one layer, the exponential appearing in [26] is now replaced by

$$[\mathcal{L}_m(d_m)]^{1/d_m} = (I + K_m d_m)^{1/d_m}. \tag{29}$$

In the right basis, where $K_m$ is diagonal, we get

$$\left( 1 \pm i k^n_m d_m \right)^{1/d_m} = \exp \left[ \frac{\ln \left( 1 \pm i k^n_m d_m \right)}{d_m} L \right]. \tag{30}$$

on the diagonal. In [26], we had $\exp(\pm i k^n_m d_m)^{1/d_m}$. The difference on the complex phase accumulated over one layer ($L = d_m$) is therefore given by

$$\Delta \Phi(d_m) = k^n_m d_m - \ln \left( 1 + i k^n_m d_m \right) / i. \tag{31}$$

By expanding $\Delta \Phi$ in Taylor series, it is possible to show (see [26, Sec. VI]) that this error is bounded by

$$|\Delta \Phi| < \frac{3}{2} \frac{|k^n_m|^2 d_m^3}{(1 - \ln|k^n_m| d_m)^3}. \tag{32}$$

in a lossy material.

To get an estimate of this error bound, we consider a material with thickness 0.5 nm, at a wavelength of 1000 nm and a complex refractive index on the order of 1, both for the real and imaginary parts. This leads to $|k^n_m| d_m < |n| k_0 d_m = 2.2 \pi \cdot 0.5/1000 \approx 6 \cdot 10^{-5}$, that is an absolute error $|\Delta \Phi| < 6 \cdot 10^{-5}$, and a relative error lower than $10^{-5}$. For an effective refractive index of 10, this value would be reduced to 0.1.

At the moment, this error is small in view of the precision of the measurements performed on single-layer materials, but it could become important, especially at shorter wavelengths.

### IV. ENERGY CONSERVATION

The layer matrix [12] is conservative when $k^n_m$ is a real quantity. However its first order expansion in $\Phi$ could be criticized because it violates flux conservation for insulators [21].

While this is true, from an algebraic perspective, this is fortunately not the case if the calculation is limited to the first order. To understand the origin of the problem, we consider [11] in the case where $k^n_m$ is real. The interface matrices are not concerned with the Taylor expansion, and the flux violation can therefore not come from these matrices. The propagation matrix provides $r = 0$ and $t = \exp(i k^n_m d_m)$. Flux conservation is therefore verified if $k^n_m$ is real. However, when $t$ is expanded to first order, we get $t = 1 + i k^n_m d_m$ so that $|t|^2 = 1 + |k^n_m d_m|^2$ showing that the flux is no more conserved. The error is of second order, which is negligible for typical values of $\Phi = k^n_m d_m$, as shown in Sec. III C and makes sense from an analytical point of view.

Though this is not problematic, it would be more comfortable to work with an approximation that ensure flux conservation. One way to achieve this is to rewrite the imaginary exponential before proceeding to the Taylor expansion. Writing

$$\exp(i \Phi) = \frac{\exp(i \Phi/2)}{\exp(i \Phi/2)} = 1 + i \tan(\Phi/2) / 1 - i \tan(\Phi/2), \tag{33}$$

provides the expansion

$$\exp(i \Phi) = 1 + i \Phi/2 + \Theta(\Phi^3) \tag{34}$$

where $\Theta(\Phi^3)$
that is algebraically conservative when Φ is a real quantity, as
\[
\left| \frac{1 + i\Phi}{2} \right|^2 = 1 + \Phi^2/4 = 1. \tag{35}
\]

We will use this higher order approximation in Sec. VII to build the layer matrix, and in Sec. IX to fit the model to experimental data.

V. LAYER MODEL TO BOUNDARY CONDITIONS

We are now ready to calculate the boundary conditions associated to a layer matrix and to build the layer matrix from the boundary conditions, in a way that will directly allow to calculate the matrix of stacked 2D materials.

When presenting anisotropic boundary conditions at an interface that contains surface currents, we have seen that the surface polarization terms are associated to the quantities ϕ and ψ in Eqs. (1) and (2). These functions can easily be calculated from the layer transfer matrix (12) with the expansion of (34) that is rewritten to separate the even and the odd parts in Φ
\[
\cos(Φ) + i\sin(Φ) \approx \frac{4 - Φ^2}{4 + Φ^2} + \frac{4Φ}{4 + Φ^2}. \tag{36}
\]

This provides (see [26 Sec. IIA]) the transmittance and reflectance:
\[
T_L = 1 + \frac{iΦ_m/α_m}{2 - iΦ_m/α_m} + \frac{iα_mΦ_m}{2 - iα_mΦ_m}, \tag{37}
\]
\[
R_L = \frac{iα_mΦ_m}{2 - iα_mΦ_m} - \frac{iΦ_m/α_m}{2 - iΦ_m/α_m}, \tag{38}
\]
from which one can calculate
\[
T_L \left( \frac{1}{T_L} - \frac{R_L}{T_L} - 1 \right) = -\frac{2iΦ_m/α_m}{2 - iΦ_m/α_m}, \tag{39}
\]
\[
T_L \left( \frac{1}{T_L} - \frac{R_L}{T_L} - 1 \right) = -\frac{2iα_mΦ_m}{2 - iα_mΦ_m}. \tag{40}
\]

Now comes the interesting physical interpretation. We expect that in the limit of infinitely thin layers, the right-hand side of these last equations will provide the jump conditions appearing in [1] and [2].

In a first approach, we can consider that the permittivity and the surface susceptibility are related by (23–25) so that in the limit of very small d_m, d_mexp/ε_0 ≈ ε exp and d_mexp/ε_s ≈ ξ exp. In a second approach we compute the interface matrix \( S_{ab} \) from \( L_m \) using back propagation
\[
S_{ab} = P_{ab}(-d_m/2)L_m(d_m)P_{ab}(-d_m/2), \tag{41}
\]
and we calculate \( T_L + R_L - 1 \) from this matrix to identify \( ϕ \) and \( ψ \) from the boundary conditions [1]–[2].

Both approaches provide the same results (Sec. III). This is important as it explains why the layer \( L(d_m) \) can be presented either as a continuous layer as shown on Fig. 1(center), or as current sheet surrounded by vacuum, as depicted on Fig. 1(right). This dual character of the ALM allows to connect volume parameters such as permittivity, and propagation constant to surface parameters such as surface susceptibilities.

As \( ϕ_{ab} \) and \( ψ_{ab} \) have different expressions for TE and TM configurations, they should be calculated separately. This provides
\[
ϕ_{ab}^{\parallel} = 0, \tag{42}
\]
\[
ϕ_{ab}^{\perp} = k_{xx}^0 x_s^y, \tag{43}
\]
\[
ψ_{ab}^{\parallel} = \frac{k_{xx}^0}{k_{zz}} x_s^y, \tag{44}
\]
\[
ψ_{ab}^{\perp} = \frac{k_{xx}^0}{k_{zz}} x_s^y, \tag{45}
\]
with \( x_s^y \) defined in [6].

As expected from the definition of a layer with intrinsic parameters, there is no coefficient involving parameters of the input or output medium.

Starting from boundary conditions [1] and [2], we build the interface matrix \( S_{ab} \) of a 2D layer surrounded by vacuum using (3) and the symmetry of the system leading to \( r = r', t = t' \). As detailed in [26 Sec. IIA], we get
\[
S_{ab} = I + \begin{pmatrix}
-i\psi_{xx} - i\psi_{yy} & i\psi_{zz} & i\psi_{yy} - i\psi_{xx} \\
-i\psi_{yy} + i\psi_{xx} & i\psi_{zz} & i\psi_{zz} + i\psi_{xx}
\end{pmatrix}. \tag{46}
\]

The layer matrix \( L_m \) is then built using [22] with an effective thickness \( d_m \) that can be measured through electron microscopy [4], and a first order expansion of
\[
L_m(d_m) = I + ik_z^0 d_m \begin{pmatrix}
-1 & 0 \\
0 & 1
\end{pmatrix}. \tag{47}
\]

The first-order layer matrix is
\[
L_m(d_m) = I + \begin{pmatrix}
-A & B \\
-B & A
\end{pmatrix} d_m, \tag{48}
\]
that provides, at first order, a $K_{th}$ matrix similar to \( K \), with $A = A_1 d_1 + A_2 d_2$, $B = B_1 d_1 + B_2 d_2$ and $d_h = d_1 + d_2$. Using (51)-(52) this leads to
\begin{align*}
\beta^2_{h_1} &= \frac{d_1}{d_h} \beta^2_1 + \frac{d_2}{d_h} \beta^2_2, \\
\beta^2_{h_2} &= \sum_{i=1}^{2} \frac{d_i}{d_h} \beta^2_i \left[ d_1 + d_3 \left( \varepsilon_{x,i} - \varepsilon_{x,1} \right) \right].
\end{align*}

After expanding $k_{x}^2$, $x = v, 1, 2, h$ as in (A.8) and (A.11), we get the effective permittivities of the hybrid medium
\begin{align*}
\varepsilon_{x}^h &= \frac{d_1}{d_h} \varepsilon_{x,1} + \frac{d_2}{d_h} \varepsilon_{x,2}, \\
\varepsilon_{y}^h &= \frac{d_1}{d_h} \varepsilon_{y,1} + \frac{d_2}{d_h} \varepsilon_{y,2}, \\
\varepsilon_{z}^h &= \frac{1}{d_h} \frac{1}{d_1} + \frac{1}{d_h} \frac{1}{d_2}.
\end{align*}

A. Discussion

If we expand the permittivity in terms of susceptibility, (57) and (58) take the form
\begin{align*}
1 + \chi_h &= \frac{d_1}{d_h} (1 + \chi_1) + \frac{d_2}{d_h} (1 + \chi_2), \\
d_h \chi_h &= d_1 \chi_1 + d_2 \chi_2.
\end{align*}

showing that if the surface susceptibility is $\chi^s = d \chi$, the total surface susceptibility is the sum of the individual surface susceptibilities, in which the thicknesses of the layers do not appear: $\chi^s_h = \chi^s_1 + \chi^s_2$.

We see that the order of the layers does not enter into account. This conclusion is valid within the error bound given in Sec. III.B. It corresponds to the classical vision that on a subwavelength scale, dipole contributions can be summed to model the material response.

We see here that the total dipole contribution is handled separately from the total thickness. Though we can consider pure surface contributions from the dipole response of the 2D materials, we cannot neglect the thickness of the individual layers.

However, for the out-of-plane component, it is better to introduce the volume quantity $\xi = (\varepsilon_{z} - \varepsilon_0) / 2 \varepsilon_z$ so that (59) becomes
\begin{align*}
\frac{1}{\varepsilon_0} - \frac{1}{\varepsilon_{z,h}} &= \frac{1}{\varepsilon_0} - \frac{d_1}{d_h} \frac{1}{\varepsilon_{z,1}} - \frac{d_2}{d_h} \frac{1}{\varepsilon_{z,2}}, \\
d_h \xi_h &= d_1 \xi_1 + d_2 \xi_2.
\end{align*}

showing that if we define the surface quantity $\xi^s_h = d \xi^s$, $\xi^s_1 + \xi^s_2$. This allows to extend the conclusions on $\chi^s_h$ and $\chi^s_v$ to $\xi^s_h$.

An important conclusion from (57) is that out-of-plane permittivity components do not sum in hybrid materials while the surface quantities $\xi^s$ do.

VII. HYBRID MATERIAL

It is now time to show the practical importance of the layer approach by providing effective parameters for a medium made of stacked alternating layers of 2D materials. We consider two kinds of layers characterized by two different susceptibility tensors and thicknesses (Fig. 3). The elementary layer of this hybrid material correspond to the product of two single layers

\[
\mathcal{L}_h = \mathcal{L}_1 \mathcal{L}_2
\]
\[
= \left[ I + i \begin{pmatrix} -A_1 & B_1 \\ -B_1 & A_1 \end{pmatrix} d_1 \right] \times
\left[ I + i \begin{pmatrix} -A_2 & B_2 \\ -B_2 & A_2 \end{pmatrix} d_2 \right],
\]

FIG. 3: Hybrid structure made of alternated layers of type 1 and 2. The structure is modeled as a set of hybrid layers ($h$).

\[
A = \frac{\varphi_{vv}^m + \psi_{vv}^m}{2t_{dm}} + k_z^v, \\
B = \frac{\varphi_{vv}^m - \psi_{vv}^m}{2t_{dm}}.
\]

that provides, at first order, a $K_{th}$ matrix similar to \( K \), with $A = A_1 d_1 + A_2 d_2$, $B = B_1 d_1 + B_2 d_2$ and $d_h = d_1 + d_2$. Using (51)-(52) this leads to
\begin{align*}
\beta^2_{h_1} &= \frac{d_1}{d_h} \beta^2_1 + \frac{d_2}{d_h} \beta^2_2, \\
\beta^2_{h_2} &= \sum_{i=1}^{2} \frac{d_i}{d_h} \beta^2_i \left[ d_1 + d_3 \left( \varepsilon_{x,i} - \varepsilon_{x,1} \right) \right].
\end{align*}

After expanding $k_{x}^2$, $x = v, 1, 2, h$ as in (A.8) and (A.11), we get the effective permittivities of the hybrid medium
\begin{align*}
\varepsilon_{x}^h &= \frac{d_1}{d_h} \varepsilon_{x,1} + \frac{d_2}{d_h} \varepsilon_{x,2}, \\
\varepsilon_{y}^h &= \frac{d_1}{d_h} \varepsilon_{y,1} + \frac{d_2}{d_h} \varepsilon_{y,2}, \\
\varepsilon_{z}^h &= \frac{1}{d_h} \frac{1}{d_1} + \frac{1}{d_h} \frac{1}{d_2}.
\end{align*}

A. Discussion

If we expand the permittivity in terms of susceptibility, (57) and (58) take the form
\begin{align*}
1 + \chi_h &= \frac{d_1}{d_h} (1 + \chi_1) + \frac{d_2}{d_h} (1 + \chi_2), \\
d_h \chi_h &= d_1 \chi_1 + d_2 \chi_2.
\end{align*}

showing that if the surface susceptibility is $\chi^s = d \chi$, the total surface susceptibility is the sum of the individual surface susceptibilities, in which the thicknesses of the layers do not appear: $\chi^s_h = \chi^s_1 + \chi^s_2$.

We see that the order of the layers does not enter into account. This conclusion is valid within the error bound given in Sec. III.B. It corresponds to the classical vision that on a subwavelength scale, dipole contributions can be summed to model the material response.

We see here that the total dipole contribution is handled separately from the total thickness. Though we can consider pure surface contributions from the dipole response of the 2D materials, we cannot neglect the thickness of the individual layers.

However, for the out-of-plane component, it is better to introduce the volume quantity $\xi = (\varepsilon_{z} - \varepsilon_0) / 2 \varepsilon_z$ so that (59) becomes
\begin{align*}
\frac{1}{\varepsilon_0} - \frac{1}{\varepsilon_{z,h}} &= \frac{1}{\varepsilon_0} - \frac{d_1}{d_h} \frac{1}{\varepsilon_{z,1}} - \frac{d_2}{d_h} \frac{1}{\varepsilon_{z,2}}, \\
d_h \xi_h &= d_1 \xi_1 + d_2 \xi_2.
\end{align*}

showing that if we define the surface quantity $\xi^s_h = d \xi^s$, $\xi^s_1 + \xi^s_2$. This allows to extend the conclusions on $\chi^s_h$ and $\chi^s_v$ to $\xi^s_h$.

An important conclusion from (57) is that out-of-plane permittivity components do not sum in hybrid materials while the surface quantities $\xi^s$ do.
VIII. INTRINSIC SURFACE QUANTITIES

In Sec. [3C] we have seen that in the particular case where a layer is surrounded by vacuum, \( \chi_z^i \) as defined in [20] is equal to \( \xi_z^i \) defined in [9]. This will not be the case anymore if the surrounding material differs from vacuum. As a particular case, we consider a layer of isotropic material \( m \) immersed in the same material \( m \). Due to the isotropy of medium \( m \), we have for the volume quantities

\[
\chi_x = \frac{\varepsilon_x}{\varepsilon_0} - 1 = \frac{\varepsilon_x}{\varepsilon_0} - 1 = \chi_z.
\]
\[
\xi_z = \frac{\varepsilon_z - \varepsilon_0}{\varepsilon_z} = \frac{\varepsilon_z - \varepsilon_0}{1 + \chi_z}.
\]

If we define the surface quantities \( \chi_z^i = \lim_{d \to 0} d \chi_i \), for \( i = x, y, z \), then \( \xi_z^i = 1 \). As we have seen that the intrinsic surface quantity for the out-of-plane response is well-defined \( \chi_z^i \), this means that \( \chi_z \) is not a well-defined quantity, and it should not be used to describe intrinsic parameters.

On the other hand, we see that the unusual relation between the susceptibility and the permittivity appearing in \( \text{(25)} \) is the natural connection between \( \xi_z \) and \( \varepsilon_z \), both for the bulk quantity and the surface limit.

This shows that \( \xi_z \) is the surface quantity that should be used to describe the out-of-plane response of a surface material, while \( \chi_z \) and \( \chi_z^i \) are used for the in-plane response.

Doing so, the link between the volume susceptibility and permittivity is the same for all axes

\[
\chi_i = \frac{\varepsilon_i}{\varepsilon_0} - 1, \quad i = x, y, z,
\]

which is of particular importance in a bulk isotropic material. The surface quantities are defined through

\[
\chi_x^{m,s} = \frac{P_x}{\varepsilon_0} = \frac{d_m}{\varepsilon_m} = \frac{d_m}{\varepsilon^{m}_0} - \varepsilon_0,
\]
\[
\xi_z^{m,s} = \frac{P_z}{D_z} = \frac{d_m}{\varepsilon^{m}_0} - \varepsilon_0
\]

with \( y \) component obtained by the substitution \( x \to y \) in \( \chi_z \).

These quantities are intrinsic in that they do not depend on the parameters of the surrounding layers. To clearly understand the meaning of this, remember that we have defined a layer as being surrounded by vacuum. In practice, when we evaluate the surface quantities of a layer \( m \) on a substrate \( b \), we could do so based on the field in medium \( m \) [seen as a continuous medium, Fig. [1] center)]; in medium \( b \); or in the vacuum when the layer is seen as a current sheet surrounded by vacuum [see Fig. [1] right].

The field and displacement components that appear in \( \text{(67)} \) and \( \text{(68)} \) are continuous across a surface without surface currents. This is a direct consequence of \( \text{(A.1)} \), \( \text{(A.3)} \) and \( \text{(A.4)} \) for an interface without surface currents \( (\mathcal{P} = 0) \) which means that we can decompose the interface matrix between medium \( m \) and substrate according to \( \mathcal{J}_{\text{mb}} = \mathcal{J}_{\text{mv}} \mathcal{I}_{\text{vb}} \), and assume that these three components are the same in \( m, v \) and \( b \), confirming that the surfaces quantities \( \chi_x^{m,s}, \chi_y^{m,s} \) and \( \xi_z^{m,s} \) are intrinsic.

These three quantities can be summed directly when stacking layers.

This is especially useful to build an hybrid material, as the number of layers is limited. However, calculations in Sec. [VII] were performed in the first order limit and their precision is limited as estimated in Sec. [III E].

IX. FITTING THE MODELS TO EXPERIMENTAL DATA

Data recording optical properties of 2D materials at different angles are very limited in the literature. This is probably due to the difficulty to record them with classical setups, as described in [20] and [29]. To circumvent this difficulty, Xu [27] proposed an experimental configuration in which a 2D material is characterized in two steps: (i) on a polymer substrate; (ii) with the same polymer added on top, so that the 2D material is immersed in the polymer. We will investigate how to fit these results with the interface and the layer models.

A. Fitting with the interface model

A microscopic description of a 2D material [22] was used in [21] to fit experimental ellipsometric data taken on graphene and MoS2 immersed in a polymer.

The reflectivity \( r_{ip} \) from [27, Eq.3] is written in the notation of this article in [26, Sec. VII], [26, Eq. (156)]. This configuration can be obtained from the layer model as follows, with a indicating parameters of the polymer layer

\[
\mathcal{M} = \mathcal{I}_{av} \mathcal{L}_m(d_m) \mathcal{I}_{va} = \mathcal{I}_{av} \mathcal{I}_{vm} \mathcal{P}_m(d_m) \mathcal{I}_{mv} \mathcal{I}_{va} = \mathcal{I}_{am} \mathcal{P}_m(d_m) \mathcal{I}_{na}.
\]

As the coefficient \( \alpha \) in \( \text{(12)} \) comes from the propagation matrix, it is sufficient to replace \( \alpha_{vm} \) by \( \alpha_{am} \) in \( \text{(39)} \) and \( \text{(40)} \) to find the expression for an immersed layer of material \( m \). To go from this thick layer to the interface model, we can then let \( d_m \to 0 \) as described in Sec. [V] and detailed in [26, Sec. III]. Writing

\[
\mathcal{K}_x = \lim_{d_m \to 0} \mathcal{I}_{am} \mathcal{P}_m(d_m) \mathcal{I}_{na} = \frac{k_x^2 \varepsilon_0}{k_x^2 \chi_x^s},
\]
\[
\mathcal{K}_y = \lim_{d_m \to 0} \mathcal{I}_{am} \mathcal{I}_{na} = \frac{k_z^2}{k_z^2 \varepsilon_z^s},
\]
\[
\mathcal{K}_z = \lim_{d_m \to 0} \mathcal{P}_m(d_m) = \frac{k_x^2}{k_x^2 \varepsilon_x^s}
\]

\[\text{(73)}\]

provides the reflection coefficients

\[
R_y = \frac{i \mathcal{K}_y}{2 - i \mathcal{K}_y},
\]
\[
R_z = -\frac{i \mathcal{K}_z}{2 - i \mathcal{K}_z}.
\]

\[
\text{(75)}\]
for a 2D material immersed in medium \(a\).

A comparison of (39) and (40) with [26] Eq. (159)–[26] Eq. (163) shows that these expressions are identical if \(\kappa_i = \tau_i, \; i = x, y, z\). This occurs when

\[
\xi_z^s = \frac{\chi^s}{n^a_2}, \tag{76}
\]

and we take the same sign convention for the Fourier transform. This is illustrated on Fig. 4 where the two curves of the MRM and AIM overlap perfectly.

**B. Fitting with the layer model**

When plotted with the same values the MRM and the ALM differ strongly, as shown on Fig. 4. As the MRM fits nicely to experimental data (see [21]), this seems to favor the AIM and MRM with respect to the ALM that was presented in Sec. III.

However, quite surprisingly, the ALM can fit to the same data as the MRM, with the same accuracy. It can even do better, if it can fit the MRM model itself. The result is presented on Fig. 4 (diamonds) where the difference between the curves is extremely small, either for the reflectance curves and the ellipsometric curves. Interestingly, all reflectance curves are very similar in TE configuration. The fitting parameters used to draw curves from Fig. 4 are given in Tab. II. The parameter \(\chi^s\) is used for the MRM, while \(\xi_z^s\) is used for ALM. We use (76) to compare both models in Tab. II. The difference between the ALM and the AIM comes from the additional propagation in vacuum in AIM, and mainly affects the real part of the in-plane and of the out-of-plane susceptibilities.

While the same curve can be fitted equally well with the two models, the retrieved values for \(\chi_x\) and \(\chi_z\) are different (see Tab. II).

**FIG. 5:** Multilayer system. Schematic representation of the different models. \(a\) (green) and \(b\) (blue) denote surrounding media, \(v\) (white) represents vacuum. Red lines correspond to surface currents. The interface configurations (MRM and AIM) do not contain vacuum between the surface current and the surrounding material. In the layer model (ALM) surface currents are separated by vacuum. In other representations, a single current sheet is considered, with vacuum on one or two sides to compensate for the physical thickness.

Results on MoS\(_2\) can also be fitted successfully by the two models ([26] Fig. 1).

**X. MULTILAYER SYSTEM**

Now, we turn to the main goal which is to model multilayer systems. Using the layer matrix (12) it is straightforward to proceed in a 

Starting from a single layer, we take the N-th power to model a N-layer stack of 2D materials. This is compared on Fig. 5 to two other approaches. The interface approach (MRM, AIM) does not take into account the thickness of one layer and is therefore not scalable. The third approach consists in adding dipole contributions or surface currents in a single sheet and to surround it by vacuum or air to compensate for the total thickness. Different positions of the sheet are shown on Fig. 5 (int+vac).

The TE or TM reflectance corresponding to these 5 configurations are compared on Fig. 6 for 10 and 100 layers.

For a limited number of layers in TE configuration, the change of reflectance between the interface and the layer models is negligible with respect to the experimental precision. However, in TM configuration, there is a strong difference between the interface and the layer model. For 10

**TABLE II:** Comparison of the fitting parameters of the MRM analytical curve with either the ALM or the AIM. For the AIM & ALM, \(\chi_z^s = n^a_2 \xi_z^s\).

| Model | \(\chi^s_x\) | \(\chi^s_z\) | \(\xi^s_x\) | \(\xi^s_z\) |
|-------|-------------|-------------|-------------|-------------|
| MRM   | 1.7         | 2.58        | 0.60        | 0.11        |
| AIM   | 1.7         | 2.58        | 0.60        | 0.11        |
| ALM   | 2.04        | 2.58        | 0.94        | 0.11        |

\[\begin{array}{cccc}
\chi^s_x & \chi^s_z & \xi^s_x & \xi^s_z \\
\hline
\text{MMR} & 1.7 & 2.58 & 0.60 & 0.11 \\
\text{AIM} & 1.7 & 2.58 & 0.60 & 0.11 \\
\text{ALM} & 2.04 & 2.58 & 0.94 & 0.11 \\
\end{array}\]

\[\begin{array}{cccc}
\epsilon_x^s & \epsilon_z^s & \epsilon_x^i & \epsilon_z^i \\
\hline
\text{AIM} & 6.09 & 7.73 & 2.63 & 4.35 \\
\text{ALM} & 7.12 & 7.73 & -2.10 & 0.86 \\
\end{array}\]
FIG. 6: Reflectance of multilayer systems predicted by the different models illustrated on Fig. 5. Left: 10 layers. Right: 100 layers. Top, TE reflectance. Bottom, TM reflectance.

FIG. 7: Similar curves as in Fig. 4 for 100 layers, for a configuration where the graphene layers are on top of a substrate, with air above. The main impact of the model is on the reflectance curves.

layers, the position of the surface current within the vacuum gap has low importance. For 100 layers, the reflectance curves differ strongly, and in particular, they predict very different Brewster’s angles.

This shows the importance to select the right model when a larger number of layers is considered.

The case of a 2D material immersed in a polymer is very instructive to compare the different geometries from Fig. 5. However, in many experiments, the input medium is air, which is very close to vacuum. In that case, there is no reflection at the interface between medium $a$ and the vacuum part of the 2D layer. The reflectance due to the interface air-polymer combines with the response of the graphene sheet, leading to very similar responses on Fig. 7 for the different models.

We observe higher reflectance from the interface model than from the stacked-layer model. This is explained as follows: in the interface model, we concentrate the material response in one layer, so that the change of impedance (refractive index) at this layer is important, causing high reflection. In the stacked-layer model, the different layers make an homogeneous medium, with a smaller impedance shift at the interface, and a smaller reflection. Once light is in the homogeneous medium it is not reflected anymore.

XI. SUMMARY AND CONCLUSION

We compared different ways to model 2D materials and stacked 2D layers. We proposed to use a layer model described by intrinsic parameters. This model works well if the out-of-plane component of the polarization of the current sheet is related to $D_z$ using the displacement susceptibility $\xi_z$. The ALM leads to simple expressions of the transmission and reflection ratios that predict flux conservation in a lossless medium. It offers a complete picture where boundary conditions can be deduced from the volume parameters, or oppositely volume parameters can be obtained from boundary conditions of a single layer, showing that a first order description at this level contains all the physics. The ALM offers a dual vision of layer that can be continuous or correspond to a current sheet surrounded by vacuum. The ALM is also suitable to describe hybrid materials.

Then, we showed how the layer model evolves to the interface model when the layer is shrunk down to a zero thickness corresponding to surface currents. This is an important point to connect volume parameters to ab-initio calculation [30] that can be performed in the 2D material.

The AIM corresponds to the MRM in this limit. We have also shown that the AIM and the ALM fit equally well the experimental data for graphene and MoS2. It is therefore not possible to select the right model based on the best fit. Nonetheless, the layer model appears as the best candidate, due to its ability: (i) to describe continuous and discrete media, with a quantified error; (ii) to recover the interface model in the limit of infinitely thin layers; (iii) to describe hybrid materials, including van der Waals heterostructures; (iv) to unify the existing models, including anisotropy and out-of-plane components.

Finally, we have shown that when the input medium is air or vacuum, the layer model and the interface models provide very similar ellipsometric curves, even for 100 layers. Most results using the interface model are therefore justified as they are developed under this assumption. By means of our analysis, it is easy to extend the AIM to the ALM and to model a multilayer system.

In general, the TE configuration is well modelled by a dipole sum approach and this explains why most experiments can be modelled without taking into account the thickness of a layer, nor its internal geometry.

Our approach shows that in the immersive configuration, which is the most useful in practice to access the different parameters of the model, the thickness of the layer is important. However, the in-plane dipole contributions can be summed. For the out-of-plane component, we have introduced the displacement susceptibility $\xi_z$ allowing to assign surface values.

We have thus provided a complete picture on the way to
model interfaces, the microscopic reaction model and the thin film approach. Intrinsic parameters have been identified, that will allow to compare measurement performed with different surrounding materials. We showed how to model hybrid materials and how to quantify the error between the discrete and the continuous approaches. This will allow to characterize the intrinsic parameters of 2D materials and to calculate the response of multilayer systems. Experimental deviations from these predictions will provide information on interactions between layers.

We believe that this work answers important questions for the community. It allows to situate the different results published so far, by distinguishing the notion of interface model and layer model. While these two models may be confused in some cases, we have shown how they differ and how to apply them.

This work focuses on materials without coupling between the TE and TM components. However, if needed, the layer approach could be extended to materials that do not verify this assumption.

Appendix: Boundary conditions for TE and TM modes

As detailed in [20], boundary conditions for a TE (⊥) incident plane wave of frequency \( \omega / (2\pi) \), and wavenumber \( \vec{k} = k_1 \vec{I}_x + k_2 \vec{I}_z \), on a 2D material located in plane \( z = 0 \), are (\( t \) stands for the transmitted fields, \( r \) for the reflected fields, and \( i \) for the incident ones)

\[
E_y^t = E_y^i + E_r^i, \quad k_z^t E_y^t = k_z^i E_y^i + k_0 \vec{\omega} \omega^2 \vec{P}_y, \quad (A.1)
\]

while those for a TM (∥) wave are

\[
D_z^t = D_z^i + D_r^i - i k_z \vec{P}_x, \quad (A.3)
\]

\[
a_i^\parallel, D_z^i = D_z^i - D_z^r + i k_z^2 \varepsilon_z^i \varepsilon_0 \vec{P}_x, \quad (A.4)
\]

with \( \vec{P}_x \), the surface polarization field.

For symmetry reasons, it is most of the time possible to assume that the TE and the TM waves are not coupled by the surface polarization term, i.e. \( \chi_{z\parallel} = \chi_{z\perp} = \chi_{x\parallel} = \chi_{x\perp} = 0 \). In this case, Eqs. (A.1) and (A.3); and (A.2) and (A.4) write respectively

\[
t - r - 1 = i \psi_{tt}, \quad (A.5)
\]

\[
t_{it} + r - 1 = i \psi_{ti}, \quad (A.6)
\]

with

\[
t_{\parallel} = E_{y\parallel}^t / E_{y\parallel}^i, \quad r_{\parallel} = E_{y\parallel}^r / E_{y\parallel}^i, \quad (A.7)
\]

\[
a_{i\parallel} = \frac{k_{z\parallel}^i}{k_{z\parallel}^t}, \quad k_{z\perp}^i = \frac{\varepsilon_z^i}{\varepsilon_0^t} - k_z^2, \quad (A.8)
\]

\[
q_{i\parallel}^t = 0, \quad \psi_{ti} = \varepsilon_z^i \left( \frac{k_{z\parallel}^i}{k_{z\parallel}^t} \right) \vec{P}_y, \quad (A.9)
\]

\[
t_{\perp} = D_{z\parallel}^t / D_{z\parallel}^i, \quad r_{\perp} = D_{z\parallel}^r / D_{z\parallel}^i, \quad (A.10)
\]

\[
a_{i\perp} = \frac{k_{z\perp}^i}{k_{z\perp}^t}, \quad k_{z\parallel}^i = \frac{\varepsilon_z^i}{\varepsilon_0^t} - k_z^2, \quad (A.11)
\]

\[
\psi_{ti} = \frac{k_{z\parallel}^i}{k_{z\parallel}^t} \left( \frac{\varepsilon_z^i}{\varepsilon_0^t} \right) D_{z\parallel}^i, \quad (A.12)
\]

where \( k_0^2 = \omega^2 / c^2 \), with \( c \) the speed of light in a vacuum and \( \varepsilon_0 \) the vacuum permittivity.

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