The classical optical response of a bilayer crystal
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We extend the recently developed classical theory for the optical response of a single-layer crystal to bilayers. We account for the interaction between the two atomic planes and the multiple reflections inside the crystals. We show how to define a global susceptibility meaningful for the bilayer crystal and how its expression varies compared to the single-layer case. We compute both the local and the macroscopic fields which allow us for a direct comparison with experimental data.

INTRODUCTION

A two-dimensional (2D) atomic crystal is a single plane of atoms or molecules whose properties are significantly different from those of its three-dimensional (3D) precursor. This is true from a thermodynamic point of view and it becomes impressive when we consider electronics properties. Charge carriers in graphene are massless Dirac fermions [1]. A transition metal dichalcogenide monolayer is a direct band-gap semiconductors while bilayer, three-layer and multi-layer crystals are indirect band-gap semiconductors [2]. These single-layer crystals have promise for a large number of applications [3] because they are stable under ambient conditions and, although their reduced dimensionality, they are truly macroscopic objects [4].

In spite of their atomic layer thickness, these materials exhibit strong light-matter interaction [5, 6]. It was a surprising discovery that 2D crystal monolayers, deposited on suitable substrates, produce an optical contrast of up to several percent at specific wavelengths, making them easily visible [6, 7]. The comprehension of this phenomenon took some time for a proper theoretical description. The first analysis treated the single-layer crystal as a slab with an effective thickness [6]. Only few years later the adoption of the surface-current model allowed for a completely satisfactory analysis of the optical experiments on these crystals [8–12].

The optical response of a single-layer crystal provides direct access to its electronic properties via its macroscopic surface susceptibility and surface conductivity [12–16]. Recently a classical description of a 2D crystal has connected these macroscopic quantities to the microscopic atomic polarizability through the Clausius-Mossotti-Lorenz-Lorentz relations. First, a microscopic approach has shown that retardation effects are very relevant for the optical properties of these crystals [17]. Then, the computation of the macroscopic field has required the advanced potential solutions of the inhomogeneous Maxwell’s equations, via the radiation-reaction electric field [18].

The first successful technique to produce two-dimensional materials was exfoliation [1]. Now other growth methods are available, such as chemical vapor deposition [19]. All these experimental techniques are able to produce 2D crystals with different numbers of layers starting from single-layer materials, to bilayers, three-layers and up to the bulk. Optical contrast experiments are able to distinguish in between the number of constituent planes of a 2D crystal [20], but a proper theoretical analysis is missing already for the bilayer case.

In this paper, we aim to extend the complete classical physical picture that has been developed for the optical response of a monolayer crystal [17] to a bilayer material. In particular, we will address the following questions, how does the interaction between the two planes of a bilayer crystal influence its optical properties? How does this interaction scale with the distance between the atomic planes? Can we still use a surface susceptibility to describe a bilayer crystal or do we need to introduce a volume susceptibility?

CLASSICAL MODEL OF RADIATING BILAYER 2D CRYSTALS

We consider an insulating free-standing bilayer 2D crystal formed by \( N \) atoms per unit area placed on two...
2D Bravais lattices with lattice spacing \( a \) (Fig. 1). The two atomic planes are separated by a distance \( d \) and each atom has a polarizability \( \alpha \). A linearly polarized (along the \( x \)-direction) electromagnetic plane wave is incident on this 2D crystal with an harmonic time dependence \( e^{i\omega t} \). For simplicity normal incidence is assumed and the crystal is initially supposed to be in the vacuum \((n_1 = n_2 = 1)\). As a consequence of electromagnetic excitation, the atoms of the crystal act as oscillating dipoles

\[
\vec{p}_i(t) = \alpha_0 \vec{E}^{(1)}_{loc} e^{i\omega t}, \quad \vec{p}_2(t) = \alpha_0 \vec{E}^{(2)}_{loc} e^{i\omega t} \tag{1}
\]

where \( \epsilon_0 \) is the vacuum permittivity, \( \vec{p}_1, \vec{p}_2, \vec{E}^{(1)}_{loc}, \vec{E}^{(2)}_{loc} \) are respectively the induced dipole moments and the local fields in the first and the second layer. The superposition principle provides \( \vec{E}^{(1)}_{loc} \) and \( \vec{E}^{(2)}_{loc} \)

\[
\vec{E}^{(1)}_{loc} = \vec{E}_t e^{i\omega t} + \sum_{(m,n)}' \vec{E}^{(1)}_{n,m}(t) + \sum_{(m,n)}'' \vec{E}^{(2)}_{n,m}(t) \tag{2}
\]

\[
\vec{E}^{(2)}_{loc} = \vec{E}_t e^{i\omega (t-d/c)} + \sum_{(m,n)}' \vec{E}^{(2)}_{n,m}(t) + \sum_{(m,n)}'' \vec{E}^{(1)}_{n,m}(t)
\]

where \( \vec{E}_t \) is the incident electric field and the sums \( \sum' \) count the contributions coming from all the other dipoles of the same layer, while \( \sum'' \) those coming from all the dipoles of the other layer. The expression of the dipole fields is

\[
\vec{E}^{(i)}_{n,m}(t) = \frac{1}{4\pi \epsilon_0 r^3} \left( 3 (\vec{p}_i \cdot \hat{r}) \hat{r} - \vec{p}_i - \frac{\vec{p}_i \times \hat{r}}{c^2} \right) \tag{3}
\]

where

\[
\vec{p}_i = \vec{p}_i(t - \frac{r}{c}) = \vec{p}_i e^{i(\omega t - kr)}
\]

\[
\vec{p}_i = \vec{p}_i(t - \frac{r}{c}) + \vec{p}_i e^{i\omega t}(1 + ikr)
\]

with \( i = 1, 2 \) and where \((n,m)\) label the lattice sites located at \( r \equiv r_{n,m} \).

We will first develop a microscopic theory to compute the local fields \( \vec{E}^{(1)}_{loc}, \vec{E}^{(2)}_{loc} \). Afterwards we will consider the macroscopic theory to obtain first the polarization

\[
\vec{P} = \frac{\vec{P}_1 + \vec{P}_2}{d} = \frac{N}{d} (\vec{p}_1 + \vec{p}_2) \tag{4}
\]

and then the macroscopic electric field \( \vec{E} \) and the electric susceptibility \( \chi \) through

\[
\vec{P} = \epsilon_0 \chi \vec{E} \tag{5}
\]

**MICROSCOPIC THEORY**

We perform the sums in Eqs. (2) by dividing the components parallel to the crystal planes from those perpendicular

\[
\sum' \vec{E}^{(i)}_\parallel (n,m) = \frac{\alpha}{4\pi} \vec{E}^{(i)}_{loc\parallel} \tag{6}
\]

\[
\sum'' \vec{E}^{(i)}_\parallel (n,m) = \frac{\alpha}{4\pi} \vec{E}^{(i)}_{loc\parallel} \tag{7}
\]

\[
\sum' \vec{E}^{(i)}_\perp (n,m) = \frac{\alpha}{4\pi} \vec{E}^{(i)}_{loc\perp} \tag{8}
\]

\[
\sum'' \vec{E}^{(i)}_\perp (n,m) = \frac{\alpha}{4\pi} \vec{E}^{(i)}_{loc\perp} \tag{9}
\]

One can easily find that \( \vec{E}^{(i)}_{loc\perp} = 0 \) for a normally incident electromagnetic wave.

**Square and triangular lattice**

For the parallel components, we obtain numerical results consistent with the following expressions for the local fields

\[
E^{(1)}_{loc} = E_i + \frac{\alpha}{4\pi a e^3} \left( C_0 + i C_1 k a e^{-ikd} \right) E^{(1)}_{loc} + \left( C_0 + i C_1 k a e^{-ikd} \right) E^{(2)}_{loc} \tag{10}
\]

\[
E^{(2)}_{loc} = E_i e^{-ikd} + \frac{\alpha}{4\pi a e^3} \left( C_0 + i C_1 k a e^{-ikd} \right) E^{(2)}_{loc} + \left( C_0 + i C_1 k a e^{-ikd} \right) E^{(1)}_{loc} \tag{11}
\]

where the terms proportional to \( E^{(1)}_{loc} \) in eq. (10) and to \( E^{(2)}_{loc} \) in eq. (11) come from the sums \( \sum' \) in eq. (2) and have already been computed in ref. [12]. The terms proportional to \( E^{(2)}_{loc} \) in eq. (10) and to \( E^{(1)}_{loc} \) in eq. (11) come from the sums \( \sum'' \) in eq. (2).
For both the square and the triangular lattice we find that $C_1 = -2\pi N a^2$. For the square lattice we have

$$C_0 = \sum_{(m,n)\neq(0,0)} \frac{1}{2(n^2 + m^2)^{\frac{3}{2}}} \approx 4.517$$

$$C_d = \sum_{(m,n)} \frac{(n^2 + m^2 - 2 d^2/a^2)^{\frac{3}{2}}}{2(n^2 + m^2 + d^2/a^2)^{\frac{3}{2}}}$$

(12)

where $C_0 = 2\zeta(3/2)\beta(3/2)$, with $\zeta(s) = \sum_{n=1}^{\infty} 1/n^s$ the Riemann zeta function and $\beta(s) = \sum_{n=0}^{\infty} (-1)^n/(2n+1)^s$ the Dirichlet beta function [17]. For the triangular lattice

$$C_0 = \sum_{(m,n)\neq(0,0)} \frac{1}{2(n^2 + nm + m^2)^{\frac{3}{2}}} \approx 5.517$$

$$C_d = \sum_{(m,n)} \frac{(n^2 + nm + m^2 - 2 d^2/a^2)^{\frac{3}{2}}}{2(n^2 + nm + m^2 + d^2/a^2)^{\frac{3}{2}}}$$

(13)

Also in this case $C_0$ can be written in terms of special functions, $C_0 = 3\zeta(3/2)L(3/2, \chi_3)$, with $L(s, \chi_n)$ are Dirichlet L-series [17].

Honeycomb lattice

Let us consider a special case of bipartite lattice, where there are two different atoms in the unit cell for the single monolayers [21]. In this case we have to generalize Eqs. (2) for the local fields in the bilayer, introducing four terms, $E_{loc}^{(i,j)}$, where $i = 1, 2$ denotes the layers and $j = 1, 2$ labels the species of atoms with two different polarizabilities $\alpha_1$ and $\alpha_2$. For the structure depicted in Fig. 2 as in the case of the hexagonal boron nitride, Eqs. (10), (11) should be modified as it follows

$$E_{loc}^{(1,1)} = E_i + \frac{1}{4\pi a^3} \left[ \alpha_1 (C_0(1) + i C_1 k a) E_{loc}^{(1,1)} + \right.$$  
$$+ \alpha_2 (C_d(1) + i C_1 k a) E_{loc}^{(1,1)} +$$  
$$+ \alpha_2 (C_0(2) + i C_1 k a) E_{loc}^{(1,2)} +$$  
$$+ \alpha_1 (C_d(2) + i C_1 k a) e^{-ikd} E_{loc}^{(2,1)} \right]$$  

(14)

$$E_{loc}^{(1,2)} = E_i + \frac{1}{4\pi a^3} \left[ \alpha_2 (C_0(1) + i C_1 k a) E_{loc}^{(1,2)} + \right.$$  
$$+ \alpha_1 (C_0(2) + i C_1 k a) E_{loc}^{(1,1)} +$$  
$$+ \alpha_2 (C_d(2) + i C_1 k a) E_{loc}^{(2,2)} +$$  
$$+ \alpha_1 (C_d(1) + i C_1 k a) e^{-ikd} E_{loc}^{(2,1)} \right]$$  

(15)

$$E_{loc}^{(2,1)} = E_i e^{-ikd} + \frac{1}{4\pi a^3} \left[ \alpha_1 (C_0(1) + i C_1 k a) E_{loc}^{(2,1)} + \right.$$  
$$+ \alpha_2 (C_0(2) + i C_1 k a) E_{loc}^{(2,2)} +$$  
$$+ \alpha_1 (C_d(2) + i C_1 k a) e^{-ikd} E_{loc}^{(1,2)} +$$  
$$+ \alpha_2 (C_d(1) + i C_1 k a) e^{-ikd} E_{loc}^{(1,1)} \right]$$  

(16)

$$E_{loc}^{(2,2)} = E_i e^{-ikd} + \frac{1}{4\pi a^3} \left[ \alpha_2 (C_0(1) + i C_1 k a) E_{loc}^{(2,2)} + \right.$$  
$$+ \alpha_1 (C_0(2) + i C_1 k a) E_{loc}^{(2,1)} +$$  
$$+ \alpha_2 (C_d(2) + i C_1 k a) E_{loc}^{(1,2)} +$$  
$$+ \alpha_1 (C_d(1) + i C_1 k a) E_{loc}^{(1,1)} \right]$$  

(17)

For $\alpha_1 = \alpha_2$, we can identify $E^{(1,1)}$ with $E^{(1,2)}$ and $E^{(2,2)}$, reducing to Eqs. (10), (11). The coefficients are $C_1 = -2\pi N a^2$ and

$$C_0^{(1)} = \sum_{(m,n)\neq(0,0)} \frac{1}{2(n^2 + nm + m^2)^{\frac{3}{2}}} \approx 5.517$$

$$C_0^{(2)} = \sum_{(m,n)} \frac{1}{2(n^2 + nm + m^2 + n + 1/2)^{\frac{3}{2}}} \approx 11.575$$

$$C_d^{(1)} = \sum_{(m,n)} \frac{(n^2 + nm + m^2 - 2 d^2/a^2)^{\frac{3}{2}}}{2(n^2 + nm + m^2 + d^2/a^2)^{\frac{3}{2}}}$$

$$C_d^{(2)} = \sum_{(m,n)} \frac{(n^2 + nm + m^2 + n + 1/3 - 2 d^2/a^2)^{\frac{3}{2}}}{2(n^2 + nm + m^2 + n + 1/3 + d^2/a^2)^{\frac{3}{2}}}$$

For $d/a = 4/3$, as in the case of the hexagonal boron nitride, we get $C_0^{(1)} \approx -0.010$ and $C_0^{(2)} \approx 0.005$. Notice that $C_0^{(1)}$, $C_d^{(1)}$, and $C_1$ are the same of the triangular lattice.

Dependance of the interaction of the atomic planes on the distance $d$

Apart from the phase factor $e^{-ikd}$ that is due to the propagation of the electromagnetic radiation in vacuum, the only term in Eqs. (10) and (11) (or in Eqs. (14)-(17) that depends on the distance $d$ between the two atomic planes is $C_d (C_d^{(1)}, C_d^{(2)})$. We interpret it as a coefficient describing the interaction between the two atomic planes.
Its dependence on the distance $d$ (in units of $a$) is shown in Fig. 3. In all the cases the form of $C_d$, for $d$ large enough (Fig. 3), fits well with the expression

$$C_d \approx A_t \exp \left(-\frac{d}{\lambda_t}\right)$$

where $A_t$ and $\lambda_t$ depend on the lattice, $A_t < 0$ for square lattice and triangular lattice (for $C_d(1)$), while it is $A_t > 0$ in the case of $C_d(2)$ in the honeycomb lattice (see Fig. 3 where the values of $A_t$ and $\lambda_t$ in the three cases are reported). From eq. (18) and Fig. 3 one can see that, as soon as $d$ far exceeds $a$, $C_d$ becomes negligible. The points in Fig. 3 are obtained by finite size scaling as shown in Fig. 4 where the convergence of the sum for $C_d(1)$ at $d \approx 1.333$ (useful for hBN) is reported as an example. Even if the two layers are so far apart that we can safely assume $C_d = 0$, the local fields do not reduce to those of the one monolayer because the radiating fields from the first layer influence the other and vice versa.

**MACROSCOPIC THEORY**

We pass from the local fields to the macroscopic fields by using an approach similar to the one developed in \[18, 22\]. The macroscopic fields to be computed are the reflected and the transmitted fields $E_r$, $E_t$, plus the resultant of all the positive (negative) traveling waves between the two planes $E_+$ ($E_-$). These fields must obey the boundary conditions.

**Square and triangular lattice**

*Non-interacting case*

We note that the macroscopic surface current on the first (second) atomic plane is given by \[18, 22\]

$$i\frac{k}{\eta}N\alpha E_{loc}^{(i)} = i\omega P_i \quad (i = 1, 2)$$

For simplicity we first consider the case of a distance $d$ in between the two atomic planes big enough to have $C_d = 0$. The boundary conditions link together the macroscopic and the microscopic fields.

$$E_i + E_r = E_+ + E_-$$

$$E_i + E_r = E_{loc}^{(i)} \left(1 - \frac{\alpha C_0}{4\pi a^3}\right)$$

$$H_i - H_r = H_+ - H_- + i\frac{k}{\eta}N\alpha E_{loc}^{(i)} = H_+ - H_- + i\omega P_i$$

$$E_+ e^{-ikd} + E_- e^{ikd} = E_t$$

$$E_t = E_{loc}^{(2)} \left(1 - \frac{\alpha C_0}{4\pi a^3}\right)$$

$$H_+ e^{-ikd} - H_- e^{ikd} = H_t + i\frac{k}{\eta}N\alpha E_{loc}^{(2)} = H_t + i\omega P_2$$

The relation between $\vec{E}$ and $\vec{H}$ is $\eta \vec{H} = \hat{s} \wedge \vec{E}$, $\hat{s}$ is the unit vector along the propagation direction and $\eta$ is the impedance of vacuum. We have 6 equations and 6 unknown variables: $E_r, E_t, E_+, E_-, E_{loc}^{(1)}, E_{loc}^{(2)}$. This approach is self-consistent beacuse the solutions for $E_{loc}^{(1)}$
and $E_{loc}^{(2)}$ are identical to those provided by the microscopic equations (10), (11) with $C_d = 0$. The microscopic field in the first layer is naturally given by $E_i + E_r$ and in the second layer by $E_t$.

**Interacting case**

In the case of interacting atomic planes ($C_d \neq 0$) the system of eqs. (10) changes because (eqs. (10), (11)) the field applied to one plane induces a local field and hence a polarization also on the other plane.

$$E_i + E_r = E_+ + E_-$$

$$E_i + E_r = E_{loc}^{(1)}(1 - \frac{\alpha C_0}{4\pi a^3}) - E_{loc}^{(2)} \alpha C_d$$

$$H_i - H_r = H_+ - H_- + i \frac{k}{\eta} N \alpha E_{loc}^{(1)}$$

$$E_+ e^{-ikd} + E_- e^{ikd} = E_i;$$

$$E_t = E_{loc}^{(2)}(1 - \frac{\alpha C_0}{4\pi a^3}) - E_{loc}^{(1)} \alpha C_d$$

$$H_+ e^{-ikd} - H_- e^{ikd} = H_i + i \frac{k}{\eta} N \alpha E_{loc}^{(2)} = H_i + i \omega P_2$$

For $C_d = 0$ we recover the non-interacting case. Also for the interacting case self-consistency with the microscopic equations (10), (11) is verified.

**Honeycomb lattice**

We note that the macroscopic surface current on the first (second) atomic plane is given by $i \frac{k}{\eta} N (\alpha_1 E_{loc}^{(1,1)} + \alpha_2 E_{loc}^{(1,2)}) = i \omega P_i$ (i = 1, 2). In this case we have 8 equations and 8 unknown variables: $E_r$, $E_t$, $E_+$, $E_-$, $E_{loc}^{(1)}$, $E_{loc}^{(2)}$, $E^{(1,1)}$, $E^{(1,2)}$. These equations are reported in appendix I. The solutions for the local fields are self-consistent with the solutions of eqs. (10), (11).

**THE SUSCEPTIBILITY OF AN INSULATING BILAYER CRYSTAL**

From eq. (10) we have

$$\chi = \frac{P_{1x} + P_{2x}}{d \epsilon_0 (E_i + E_r + E_t)}. \quad (21)$$

For the square and the triangular lattice

$$\chi = \frac{4 \pi a^3 N \alpha}{4 \pi a^3 - (C_0 + C_d) d \alpha}. \quad (22)$$

For the honeycomb lattice

$$\chi = \frac{N (\alpha_1 + \alpha_2 - \alpha_1 \alpha_2 (c_0^{(1)} - c_0^{(2)}) - c_i^{(1)} + c_i^{(2)}))}{1 - (c_0^{(1)} + c_0^{(2)}) (\alpha_1 + \alpha_2) + \alpha_1 \alpha_2 ((c_0^{(1)} + c_0^{(2)})^2 - (c_i^{(1)} + c_i^{(2)})^2)} \quad (23)$$

Our calculations indicate that in the interacting case it is no more meaningful to provide a $\chi_s$ for each single plane (we would obtain different results for the two planes), but only a global $\chi$. For the non-interacting case this is of course still possible

$$\chi_s = \frac{P_{1x}}{\epsilon_0 (E_i + E_r)} = \frac{P_{2x}}{\epsilon_0 E_t} = \frac{P_{1x} + P_{2x}}{\epsilon_0 (E_i + E_r + E_t)} = \chi d \quad (24)$$

For square and triangular lattices

$$\chi_s = \frac{4 \pi a^3 N \alpha}{4 \pi a^3 - C_0 \alpha} \quad (25)$$

while for the honeycomb lattice

$$\chi_s = \frac{N \left( \alpha_1 + \alpha_2 - \frac{\alpha_1 \alpha_2 (c_0^{(1)} - c_0^{(2)})}{4 \pi a^3} \right)}{1 - \frac{(c_0^{(1)} + c_0^{(2)}) \alpha_1}{4 \pi a^3}} \quad (27)$$

For the special case of square lattices ($N = 1/a^2$) and $d = a$ we obtain

$$\chi = \frac{N \alpha}{1 - \frac{(C_0 + C_d) N \alpha}{4 \pi}} \quad (28)$$

where $N = 1/a^3$. As expected $\chi$ is closer than $\chi_s/a$ to the susceptibility of the bulk $\chi_{3D} = N \alpha/(1 - N \alpha/3)$. Indeed, for square lattice, $C_0 \approx 4.517$ and $C_d = -0.164$ so that $(C_0 + C_d)/\pi \approx 0.346$, very close to 1/3.

In the bulk, coupling a layer with at least the two nearest neighboring ones, one could naively expect to have $(C_0 + 2C_d)/4\pi$, which is even closer to 1/3, the 3D factor.

**THE FRESNEL COEFFICIENTS OF AN INSULATING BILAYER CRYSTAL**

**Free-standing bilayer crystal**

We want to express the Fresnel coefficients in term of $\chi$. The best way to do this is to write eqs. (10), (21) and (22) in term of $\chi$ and to solve them in this form.

$$E_i + E_r = E_+ + E_-$$

$$E_i + E_r = \frac{P}{\chi} + \frac{(P - P_i) C_F}{\epsilon_0}$$

$$H_i - H_r = H_+ - H_- + i \omega P_1$$

$$E_+ e^{-ikd} + E_- e^{ikd} = E_t;$$

$$E_t = \frac{P_2}{\chi} - \frac{(P - P_i) C_F}{\epsilon_0}$$

$$H_+ e^{-ikd} - H_- e^{ikd} = H_1 + i \omega P_2$$

These two last expressions are of course equal to the surface susceptibilities of the monolayers. Looking at the expressions and (22) we can compare the surface susceptibility of a monolayer with the susceptibility of the bilayer and we have

$$\chi_s \propto \frac{1}{d} \quad (29)$$

For the special case of square lattices ($N = 1/a^2$) and $d = a$ we obtain

$$\chi = \frac{N \alpha}{1 - \frac{(C_0 + C_d) N \alpha}{4 \pi}} \quad (30)$$

where $N = 1/a^3$. As expected $\chi$ is closer than $\chi_s/a$ to the susceptibility of the bulk $\chi_{3D} = N \alpha/(1 - N \alpha/3)$. Indeed, for square lattice, $C_0 \approx 4.517$ and $C_d = -0.164$ so that $(C_0 + C_d)/\pi \approx 0.346$, very close to 1/3.
Here $C_F$ has the dimension of the inverse of a distance. Even in the case of the honeycomb lattice (eqs. 31) we have 6 equations instead of 8. For the non-interacting case $C_F = 0 \text{ m}^{-1}$. For the interacting case, for the square and the triangular lattice

$$C_F = \frac{C_d}{4\pi a^2 N}$$  \hspace{1cm} (30)$$

while the value for the honeycomb lattice is reported in appendix II. Defining $r_s = E_r/E_i$, $t_s = E_t/E_i$ as the reflection and the transmission coefficients, the non-interacting case appears as a natural extension of the expressions of $\eta_1 (\omega)$ and $\eta_2 (\omega)$ obtained in ref. [12] where the surface susceptibility must be a free-standing single layer crystal (provided by formula (2) of ref. [12] where the surface susceptibility must be replaced with $\chi \cdot d$). For the interacting case we find

$$r_s = A + D; \quad t_s = B - D$$  \hspace{1cm} (33)$$

where $A$ and $B$ are respectively equal to (31) and (32), and $D$ is given by

$$D = \frac{2kd(e^{ikd} - 1)^2}{e^{ikd}(kd - 2i) - k^2 \chi} \cdot \frac{dC_F}{-ikd + e^{ikd}(4C_Fd\chi + ikd\chi + 2)}.$$  \hspace{1cm} (34)$$

As expected, due to the interaction, the Fresnel coefficients now depend explicitly also on $C_F$. This occurs only for terms at the order of $k^3 d^3$ or bigger since their Taylor expansions

$$r_s = -i\chi kd - (1 + \chi)k^2 d^2 + O(k^3 d^3)$$  \hspace{1cm} (35)$$

$$t_s = 1 - i(1 + \chi)kd - \frac{1}{2} (1 + 2\chi + 2^2)k^2 d^2 + O(k^3 d^3)$$

are the same for the non-interacting and the interacting case up to the second order, apart from the different expression of $\chi$ in the two cases.

### Bilayer crystal on a substrate

#### Semi-infinite substrate

We consider now the case of a bilayer crystal deposited on a homogeneous transparent medium ($n_1 = n_2$) which fills the half-space below it. As it was done in ref. 21 we assume that we can neglect the interaction of the 2D crystal with the substrate. With respect to eqs. 29 only the relation between $\hat{E}$ and $\hat{H}$ in the transmitted waves changes

$$\frac{n}{n_1} \hat{H}_t = \hat{s}_t \wedge \hat{E}_t;$$  \hspace{1cm} (36)$$

### Stratified substrate

For comparison with experimental data it is also useful to consider the case of a bilayer deposited on a stratified medium (fig. 1, $n_1 \neq n_2$)

$$E_i + E_r = E_i + E_{\pm}$$  \hspace{1cm} (37)$$

$$E_i + E_r = \frac{P_1}{\chi d \epsilon_0} + \frac{(P_1 - P_2)C_F}{\epsilon_0}$$

$$H_i + H_r = H_i + H_{\pm} + i \omega P_1$$

$$E_{1+} + E_{1-} = \frac{P_2}{\chi d \epsilon_0} - \frac{(P_1 - P_2)C_F}{\epsilon_0}$$

$$H_{1+} + H_{1-} = H_{1+} + H_{1-} + i \omega P_2$$

$$E_{1+}e^{i\beta} + E_{1-}e^{-i\beta} = E_t; \quad H_{1+}e^{-i\beta} - H_{1-}e^{i\beta} = H_t$$

where $\beta = k n_1 h$ and $h$ is the thickness of medium 1. For the non-interacting case, the Fresnel coefficients for these two types of substrates are still provided by (31) and (32). The only difference is that for the semi-infinite substrate $r_2$ and $t_2$ must be replaced with formula (6) of ref. [12] and for the stratified substrate with formulas obtained starting from the equation system (7) of ref. [12]. The Taylor’s expansion (see appendix II) of these expressions, for the non-interacting and the interacting case, are identical up to the second order in $kd$. Only the value of $\chi$ is different in the two cases. The first order term of these expansions are the Fresnel coefficients of the substrate.

### ANALYSIS OF OPTICAL CONTRAST MEASUREMENTS

In fig. 24 the crystal structure of the bilayer hBN is reported. The dimensions of the unit cell are: $a = 0.25$ nm and $c = 0.666$ nm. The unit cell is bimolecular, each atomic layer consists of a flat network of B$_3$N$_3$ hexagons with an interplanar distance of $d = c/2$ [21]. Figure 25 shows variations of the optical contrast (for the definition of this quantity see [4]) in the spectral range 410 nm < $\lambda < 740$ nm for the monolayer and the bilayer hBN on top of a SiO$_2$/Si wafer with a nominal thickness of 290 nm. Dots are the experimental data that have been extracted from ref. 20 via software digitization. The same paper reports the theoretical fits to these experimental data based on a slab model, and it assumes the same refractive index for the monolayer and the bilayer crystals.

The value of $\chi_s$ for the monolayer has already been deduced in ref. [15]. The dash line is the best theoretical fit
assumed $\chi_s = 1.3 \cdot 10^{-9}$ m and a SiO$_2$ thickness of 290 nm. The value of the surface conductivity was estimated to be $\sigma \leq 2 \cdot 10^{-6}$ $\Omega^{-1}$, confirming that we are dealing with an insulating dielectric. The only way to improve the fit is by varying the SiO$_2$ thickness, showing that the spectral position of the optical contrast curve depends much on the substrate. Solid line is the theoretical fit for the same values of $\chi_s$ and $\sigma$ but a SiO$_2$ thickness of 270 nm. Indeed we noticed that by increasing the thickness of the substrate the optical contrast curve translates towards the infrared and new zeros (or new oscillations as a function of the wavelength) appear on the blue side.

Starting from the Fresnel coefficients deduced from eqs. (37), the best theoretical fit for the bilayer provides a $\chi = 3.34$ and a SiO$_2$ thickness of 270 nm. For the sake of completeness the theoretical fit for the nominal thickness of 290 nm is reported as a dashed line. We have

$$\chi_s = 1.3 \text{ nm} > \chi d = 1.1 \text{ nm} \quad (38)$$

**Atomic polarizabilities**

Having both the $\chi_s$ from the optical contrast measurements of the monolayer [13, 20] and the $\chi$ for the bilayer, we can try to deduce the atomic polarizabilities. From the equations

$$\chi_s = N\left(\alpha_1 + \alpha_2 - \frac{\alpha_1 \alpha_2 (C_0^{(1)} - C_0^{(2)})}{2\pi a^3}\right) \left(1 - \frac{C_0^{(1)} (\alpha_1 + \alpha_2)}{4\pi a^3} + \frac{\alpha_1 \alpha_2 (C_0^{(1)} - C_0^{(2)})^2}{(4\pi a^3)^2}\right) \quad (39)$$

we can now extract the values of $\alpha_1$ and $\alpha_2$ that are the only unknown parameters of these two equations. We obtain $\alpha_1 = 1.1 \cdot 10^{-24}$ cm$^{-3}$ and $\alpha_2 = 19.5 \cdot 10^{-24}$ cm$^{-3}$ that have the right order of magnitude if compared with the static calculations reported in ref. [23]. Of course, as for the 3D case, atomic polarizabilities require a full quantum dynamical approach for a proper treatment.

**CONCLUSIONS**

In this paper, we provide a complete classical description of the optical response of a bilayer crystal. The approach reported here is valid also for an all-dielectric, double-layer metasurface [24]. We compute both the local and the macroscopic fields. We find that, if the distance $d$ between the two planes far exceeds the lattice constant $a$, they can be macroscopically treated as two separated monolayers even if the two layers are coupled by the radiating fields proportional to $C_1$ (while $C_d$ in eqs. [10], [11] and $C_1^{(1)}$, $C_1^{(2)}$ in eqs. [13]-[17] can be neglected). On the other hand, for shorter distances, although it is still possible to define a surface polarization and hence a surface current for each single plane, it is meaningful to provide a volume susceptibility rather than a surface susceptibility. Remarkably, the expression for the volume susceptibility is still very simple, and depends on the interaction parameter $C_d$ (or $C_1^{(1)}$ and $C_1^{(2)}$) which rapidly decreases with $d$, exhibiting an evanescent-wave character. Interestingly, for the Fresnel coefficients the effect of the coupling between the layers in the long wavelength limit (small $k$) is fully contained in the susceptibility.

Contrarily to what was assumed in ref. [21], where a thin film model was used to fit the experimental data, a comparison of our theory with the optical contrast measurements for the monolayer and the bilayer hBN confirms that the variation of the susceptibility passing from the single-layer crystal to the double-layer crystal is experimentally addressable even in the case of an insulating material.

We think that the same approach developed in this paper can be extended to study multilayer structures up to a thickness where the bulk susceptibility is found.

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AUTHOR CONTRIBUTIONS

MM conceived the idea. Both authors discussed the physical model and wrote the paper.

APPENDIX I

Macroscopic Theory: honeycomb lattice

\[
E_i + E_r = E_+ + E_-
\]

\[
E_i + E_r = E^{(1,1)}_\text{loc} - \frac{\alpha_1(E^{(1,1)}_\text{loc} C_0^{(1)} + E^{(2,2)}_\text{loc} C_d^{(2)}) + \alpha_2(E^{(1,2)}_\text{loc} C_0^{(2)} + E^{(2,2)}_\text{loc} C_d^{(2)})}{4\pi a^3}
\]

\[
E_i + E_r = E^{(1,2)}_\text{loc} - \frac{\alpha_1(E^{(1,1)}_\text{loc} C_0^{(2)} + E^{(2,2)}_\text{loc} C_d^{(1)}) + \alpha_2(E^{(1,2)}_\text{loc} C_0^{(1)} + E^{(2,2)}_\text{loc} C_d^{(2)})}{4\pi a^3}
\]

\[
C_F = \frac{C_d^{(2)}}{4\pi a^3 N} + \frac{\Delta_d \alpha_1 \alpha_2}{8 N \pi a^3 ((\Delta_d - \Delta) \alpha_1 \alpha_2 + 2 \pi a^3 (\alpha_1 + \alpha_2))(2 \pi a^3 (\alpha_1 + \alpha_2) - (\Delta + \Delta_d) \alpha_1 \alpha_2)}
\]

Notice that, for \( \alpha_1 = \alpha_2 \), this equation reduces to \( C_F = \frac{C_1 + C_2^2}{8 \pi a^3 N} \) so that the equations for the macroscopic electric fields are simply

\[
E_i + E_r = \frac{P_1}{\chi \delta \epsilon_0} + \frac{(P_1 - P_2)(C_0^{(1)} + C_d^{(2)})}{8 \pi a^3 N \epsilon_0}
\]

\[
E_i = \frac{P_2}{\chi \delta \epsilon_0} - \frac{(P_1 - P_2)(C_0^{(1)} + C_d^{(2)})}{8 \pi a^3 N \epsilon_0}
\]

APPENDIX II

Expression of \( C_F \) for the honeycomb lattice

\[
\Delta = C_0^{(1)} - C_0^{(2)}
\]

\[
\Delta_d = C_d^{(1)} - C_d^{(2)}
\]

Calling

\[
t_s = \frac{2}{n_1 + 1} - \frac{2 i (n_1 + 1 + 2 \chi)}{(n_1 + 1)^2} k d + \frac{1 + 2 n_1 + n_1^2 + 6 \chi + 4 n_1 \chi - 2 n_1^2 \chi + 8 \chi^2}{(n_1 + 1)^3} k^2 d^2 + O(k^3 d^3)
\]

The first terms of the expansions are the Fresnel coefficients of the substrate. It is easy to verify that the same holds for a stratified substrate.
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