Light propagation in dielectric materials

Ledo Stefanini and Giancarlo Reali

Dipartimento di Ingegneria Industriale e dell’Informazione,
Università di Pavia, via Ferrata 5a, 27100 Pavia, Italia

Abstract

We present a pedagogic derivation of the electromagnetic field established in a dielectric material by an impinging external field. We consider the problem from the point of view of the physical mechanism involved at the microscopic level. The internal field emerges when the material is thought of as an assembly of atoms in vacuum, each of them being polarized by the external incident field and by the re-radiated fields of all the other polarized atoms of the material. In this way, each atom becomes itself a source of secondary radiation that adds and interferes with all the other internal fields (including the internal extension of the externally impinging field), contributing to build up the total internal field within the dielectric material as well as the externally scattered field.

This picture naturally leads to a connection between the microscopic properties of the material and its index of refraction, that describes the dielectric response to the applied electromagnetic field. Calculations also show that the incident radiation is extinguished inside the dielectric by an equal but opposite field generated by the total induced polarization currents, and is substituted by a macroscopic electric field which propagates with the new speed $c/n$. 
I. MAXWELL'S EQUATIONS AND ELECTROMAGNETIC WAVES IN DIELECTRICS.

Maxwell’s equations provide a compact and elegant derivation of why a light beam, entering a transparent dielectric material, propagates inside it with a reduced speed.

Using Maxwell’s equations (in gaussian units), it is straightforward to derive the wave equations for the electric field \( \vec{E}(\vec{r}, t) \) in vacuum (identical equations hold for the magnetic field \( \vec{B}(\vec{r}, t) \)):

\[
\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = 0, \tag{1}
\]

characterised by dispersiveless propagation with constant speed \( c \), the light speed in vacuum.

When, instead, an electromagnetic wave propagates in a dielectric material (we think of it as homogeneous, isotropic, and non magnetic, and in addition that it responds linearly to the incident radiation), a polarization current is induced given by \( \vec{J}_P = \partial \vec{P}/\partial t \), which makes Eq.(1) inhomogeneous through the appearance of a source term \( \partial \vec{J}_P/\partial t = \partial^2 \vec{P}/\partial t^2 \):

\[
\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 \vec{P}}{\partial t^2}. \tag{2}
\]

In Eq.(2), \( \vec{E}(\vec{r}, t) \) and \( \vec{P}(\vec{r}, t) \) are macroscopic fields of Maxwell’s equations in dielectric materials, connected by the constitutive relation

\[
\vec{P} = \chi \vec{E} = \frac{(n^2 - 1)}{4\pi} \vec{E}, \tag{3}
\]

where \( \chi \) and \( n \) are the frequency dependent electrical susceptibility and index of refraction of the material, which also are macroscopic parameters of the theory. Using the second of the above relations, Eq.(2) becomes

\[
\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{(n^2 - 1)}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2}. \tag{4}
\]

The source term on the right hand side of Eq.(4) reveals an interesting form since it splits into two terms. A first one is relative to a propagation with the light speed \( c \) in vacuum and it cancels the corresponding term on the left hand side. A second one re-establishes, after that cancellation, the homogeneous structure of the wave equation in the material with the field that propagates with a reduced (and dispersive) speed \( c/n \).
II. A MICROSCOPIC APPROACH OF LIGHT PROPAGATION IN DIELECTRICS.

Wave propagation in its fully general manifestation is an extremely complex phenomenon for every type of wave motion we consider. An exceptionally lucid and lively presentation of this complexity is given by Feynman in an interview by Christopher Sykes that is posted on YouTube\(^2\). This complexity invites a further investigation into wave phenomena, taking different points of view in order to understand and clarify them at a deeper level.

For example, the previous analysis of light propagation in dielectrics does little to clarify the physical mechanism involved at the microscopic level. For this deeper explanation, the principle that we must consider when a light beam enters a dielectric is the atomic nature of matter. A piece of dielectric material is a volume of vacuum space filled with a collection of atoms, each polarized and behaving like a charged harmonic oscillator\(^3\) (an oscillating dipole moment) when acted on by the external incident radiation and by the fields re-radiated by all the other oscillating dipoles of the collection.

This picture, equivalent to the treatment with Maxwell differential equations as far as the final result is concerned, is mathematically very different since the atomic description of matter in interaction with radiation naturally leads to a representation through integral equations. From their solutions we get the expression of the index of refraction \(n\) of the material (expressed in terms of the microscopic parameters of the atoms, of their density, and of the frequency of the incident field). In addition, the solutions straightforwardly include an equal but opposite field, generated by the total induced polarization currents, that extinguishes the incident field inside the material (a fact known in optics as extinction theorem\(^4,5\)), and its substitution with a new internal field propagating at a reduced speed \(c/n\), despite the fact that the incident beam and the re-radiated waves by the atoms propagate with the vacuum light speed \(c\) in the interatomic vacuum within the material.

We shall present this point of view, basing our treatment on purely classical electrodynamics, but closely following the neat and concise presentation given by M.L. Goldberger and K.M. Watson in their technical monograph on collision theory\(^6\).

Let us consider a dielectric medium (it may be glass) made of \(N\) impenetrable identical atoms, kept together by their bonding interactions and distributed randomly within a volume \(V\) of arbitrary shape with the only restriction that the radius of curvature of its boundary at each point must be large compared to the wavelength \(\lambda\) of the incident electromagnetic
radiation. We further consider \( N \) to be a large number and the atomic density \( \rho \) to be approximately constant and equal to \( N/V \) within \( V \).

The medium is placed in the path of a plane electromagnetic wave with wave vector \( \vec{k} \), whose wavelength \( \lambda = 2\pi/|\vec{k}| \) is much greater than the atomic dimensions (for example, for visible light we have that the wavelength is \( \approx 10^4 \) the atomic dimensions). We also require that the radiation frequency be significantly less than the frequency corresponding to the lowest excitation energy of the atoms (\( \omega < \omega_{\alpha\beta} = (E_\alpha - E_\beta)/\hbar \)), so that the interaction of the incident radiation with the atoms may be considered as essentially elastic. The response of the dielectric material to the impinging radiation is then associated with the scattering of light by the atoms, and we anticipate that the electromagnetic propagation through it will be described by its index of refraction.

The microscopic, local electric field (monochromatic of frequency \( \omega \), with complex time dependence \( e^{-i\omega t} \)) is the real part of \( \vec{E}_n(\vec{r}_n) \) and has the general structure of the multiple scattering equations

\[
\vec{E}_n(\vec{r}_n) = \vec{E}_0(\vec{r}_n) + \sum_{m(\neq n)=1}^{N} \vec{E}_{rad,m}(\vec{r}_n), \quad (5)
\]

where \( \vec{E}_0(\vec{r}_n) \) is the incident (complex) electric field, and \( \vec{E}_{rad,m}(\vec{r}_n) \) is the field radiated by the \( m \)-th atom, all these fields being evaluated at the position \( \vec{r}_n \) of the \( n \)-th atom in \( V \).

This is a complex formula, with the self-consistent signature of multiple scattering problems, since the electric interaction of the \( m \)-th atom at the \( n \)-th atom position, described by the second term on the right hand side of Eq.(5), is itself dependent on the field acting on the \( m \)-th atom, which has exactly the same form of Eq.(5).

Having assumed that the incident radiation wavelength is \( >> \) than the atomic dimensions, the dipole approximation applies in this case, and in the second term in Eq.(5) we may use the classical scattering formula for \( E_{rad,m}(\vec{r}_n) \), given by

\[
\vec{E}_{rad,m}(\vec{r}_n) = \nabla_n \times \left[ \vec{p}_m \times \vec{r}_{nm} \frac{1}{r_{nm}^2} \left( \frac{1}{r_{nm}^2} - ik \right) e^{ikr_{nm}} \right], \quad (6)
\]

where \( \vec{p}_m \) is the electric dipole moment of the \( m \)-th atom induced by the local field \( \vec{E}_m(\vec{r}_m) \), \( \vec{r}_{nm} = \vec{r}_n - \vec{r}_m \), and \( \nabla_n \) is the gradient operator acting only on the \( n \)-th atom coordinates.

With the assumptions of isotropic dielectric and linear response, the induced electric dipole moment of an atom is connected microscopically with the local electric field acting on it by the scalar atomic polarizability,
\[ \vec{p}_m = \alpha \vec{E}_{m}(\vec{r}_m). \] (7)

A simple classical formula for the atomic polarizability can be found using the classical charged harmonic oscillator model. An easy calculation leads to the result (for a single resonance frequency)

\[ \alpha = \alpha' + i\alpha'' = \frac{e^2/m}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2} (\omega_0^2 - \omega^2 + i\gamma\omega), \] (8)

where \( e, m, \omega_0, \gamma \) are the oscillator characteristic parameters and \( \omega \) is the frequency of the electric driving force.

We can now go over to a continuum representation of matter, replacing the sum over \( m \) in Eq. (5) with an integral over the volume \( V \) of the dielectric except for the exclusion of a sphere \( S_0 \) with atomic radius and centered at the \( n \)-th atom. Since we made the assumptions that atoms are impenetrable, we are guaranteed that only the chosen atom is excluded as required by the sum in Eq. (5). We thus obtain (in the following, \( \vec{r}_n \) and \( \vec{r}_m \) should be regarded as continuous vectorial variables)

\[ \vec{E}_n(\vec{r}_n) = \vec{E}_0(\vec{r}_n) + \rho \int_{V-S_0} d^3r_m \vec{E}_{\text{rad},m}(\vec{r}_n) \] (9)

Eq. (9), together with expressions (6) and (7), represents the integral equation for the field \( \vec{E}_n \). Its solution will give the electric field at every point inside \( V \). To do this, we proceed according to the arguments of classical macroscopic electrodynamics, defining the macroscopic field in the medium, \( \vec{E}(\vec{r}) \), as

\[ \vec{E}(\vec{r}) = \vec{E}_0(\vec{r}) + \rho \int_{V-cyl} d^3r_m \vec{E}_{\text{rad},m}(\vec{r}), \] (10)

where in this case the integral is over the volume \( V \) excluding a very small cylindrical, needle-shaped region which includes the point \( \vec{r} \) and which is parallel to \( \vec{E}(\vec{r}) \).

Subtracting (10) from (9), and using again this last equation to eliminate the local field \( \vec{E}_n(\vec{r}_n) \), we obtain

\[ \rho \int_{V-S_0} d^3r_m \vec{E}_{\text{rad},m}(\vec{r}_n) = \vec{E}(\vec{r}_n) - \vec{E}_0(\vec{r}_n) - \rho \int_{S_0-cyl} d^3r_m \vec{E}_{\text{rad},m}(\vec{r}_n). \] (11)

We thus see that the complex expression of the dipolar interaction among the atoms on the left hand side, that also appears in Eq.(9), is equal to the right hand side combination of the incident field, the internal macroscopic field, and a term that reproduces the one on the
left hand side except that the integral now extends to the (atomic-sized) volume contained between the sphere $S_0$ and the needle-shaped surface cyl that intersects it.

To evaluate this integral, we make a further simplification based on the fact that the wavelength of the incident field is $\gg$ than the atomic dimensions. This allows us to neglect the term $i k$ in Eq.\((6)\) as compared with $1/r_{nm}$ (a quasi-static approximation), and to set $e^{i k r_{nm}} = 1$. We can further consider $\vec p_m = \alpha \vec E_m(\vec r_m)$ independent of $\vec r_m$ within $S_0$, thus replacing $\vec p_m$ with $\vec p_n$. Then Eq.\((6)\) becomes for small $r_{nm}$:

$$\vec E_{rad,m}(\vec r_n) = \nabla_n \times \left( \frac{\vec p_m \times \vec r_{nm}}{r_{nm}^3} \right)$$

$$= -\nabla_m \times \left( \frac{\vec p_n \times \vec r_{nm}}{r_{nm}^3} \right)$$

$$= \nabla_m \left( \vec p_n \cdot \frac{\vec r_{nm}}{r_{nm}^3} \right)$$

(12)

In the final expression of Eq. \((12)\), we have retained only the term that gives a non-zero contribution to the integral on the right hand side of Eq. \((11)\) (see Appendix). So the volume integral can be carried out, using standard vector analysis results, transforming it to a surface integral. This vanishes on the needle-shaped surface of the cylinder, and only receives a contribution from the integration on the surface $S_0$. The result is

$$\int_{S_0-cyl} d^3 r_m \vec E_{rad,m}(\vec r_n) = \int_{S_0-cyl} d\vec a_m \left( \frac{\vec p_n \cdot \vec r_{nm}}{r_{nm}^3} \right) = -\frac{4\pi}{3} \vec p_n.$$  

(13)

Remembering the definition of the polarization vector and using Eq. \((7)\) for the electric dipole moment, we can write $\vec P(\vec r_n) = \rho \vec p_n = \rho \alpha \vec E_n(\vec r_n)$, and this can be used with the equations \((9)\), \((11)\), and \((13)\) to obtain the final result

$$\vec P(\vec r) = \rho \alpha \vec E_0(\vec r) + \rho \alpha \left[ \vec E(\vec r) - \vec E_0(\vec r) + \frac{4\pi}{3} \vec P(\vec r) \right].$$

(14)

This is the self-consistent solution of the integral equation \((9)\) we were looking for, which evidences how the microscopic response of the material translates into a macroscopic polarization, the source term for all that propagates within the dielectric and also emerges out of it (internal field, scattered field, as well as, for special geometries with flat interfaces of the dielectric material, refracted, reflected, and transmitted fields). It is also worth noting at this point that no hypotheses have been made about the directions of the incident oscillating electric field and of its wave vector, so that the obtained relation is general.

When we only consider the internal fields, it is immediately evident from \((14)\) that one of the effects of the atomic dipolar re-radiations (the square bracket in this equation) is to
cancel the incident radiation field from within the dielectric: this is the expression of the
extinction theorem.

In addition, Eq.(14) shows the content of multiple scattering of the atomic dipolar re-
radiations as they reproduce the macroscopic polarization also on the right hand side of it,
in case of dense materials. After the cancellation of the incident field, and considering that
within an isotropic material the polarization and the electric field vectors are parallel, we
obtain

\[ P = \frac{\rho \alpha}{1 - \frac{4\pi}{3} \rho \alpha} E = \frac{n^2 - 1}{4\pi} E \]  \hspace{1cm} (15)

where for the second expression the definition of index of refraction has been used. This is
the promised connection between the microscopic parameters and the index of refraction.

In case of low density materials (such as gases), we have \( \rho \alpha \ll 1 \), and Eq. (15) simplifies,
providing a simpler relation between the atomic polarizability and the index of refraction.
The equation becomes \( n^2 - 1 = 4\pi \rho \alpha \) and, since \( (n - 1) \ll 1 \) in the given approximation,
it assumes the final form

\[ n = 1 + 2\pi \rho \alpha. \]  \hspace{1cm} (16)

In this case the enhancement effect due to multiple scattering is absent.

Since Eq.(16) is a very often used expression for the refractive index in the elementary
expositions, we would like to add few more comments on it. In the theory of scattering,
when the above approximation applies, it is common to describe the scattering off a scatterer
(like an atom or, its classical counterpart, a charged harmonic oscillator) by the scattering
amplitude \( f(\vec{k}, \vec{k}') \), where the incoming and outgoing wave vectors, in the case of elastic
scattering that we considered, satisfy the relation \( |\vec{k}| = |\vec{k}'| = k \). Consider a dielectric
material in the form of a very extended but thin slab, with thickness \( L \ll 1/k \), placed in
the plane \( z = 0 \). From what already assumed, the atoms within it have a very low packaging
density (\( \rho \alpha \ll 1 \) is satisfied). On this slab a plane electromagnetic wave (assume unit
amplitude, for simplicity) is incident along the direction \( z \). The electric field computed in a
point \( z \gg L \) assumes the form of a superposition of the incident field and of the scattered
fields in \( z \) by all the atoms of the slab\[7\]

\[ E(z) = e^{ikz} + e^{ikz} \frac{2\pi i \rho L}{k} f(\vec{k}, \vec{k}) \approx e^{ikz} e^{i\frac{2\pi \rho L}{k}} f(\vec{k}, \vec{k}) \] \hspace{1cm} (17)

where in the final results appears the forward (zero angle) scattering amplitude \( f(\vec{k}, \vec{k}) \).
f(0), and the expression of the final formula derives from the the assumed approximation. But the exponent of the last exponential of Eq. (17) has a very familiar expression in elementary optics, being equal to \(i(n - 1)kL\), so that, by comparison, we get the well-known expression

\[
n = 1 + \frac{2\pi \rho}{k^2} f(0),
\]

and, lately, comparing this expression with Eq. (16), we obtain the relation between forward scattering amplitude and atomic polarizability:

\[
f(0) = k^2 \alpha.
\]

There exists a remarkable relation (of general validity for all the scattering processes, not only for light), called the optical theorem, that connects the forward scattering amplitude and the total extinction (scattering plus absorption) cross section. For the case considered of elastic scattering, this result is easily derived. Having chosen the incident radiation to be of unit amplitude, the measured intensity at a distant forward point \(z\) is proportional to

\[
|E(z)|^2 = \left| e^{i2\pi \rho \frac{L}{k} f(0)} \right|^2 = e^{-\frac{4\pi \rho \frac{L}{k} Im f(0)}} = e^{-\rho \sigma L}
\]

where the last expression only includes the scattering cross section, since scattering is the only loss mechanism that we considered during the crossing of the slab along the propagation direction of the beam. We could as well describe intensity reduction in the forward direction as interference effect between the incoming beam and the scattered beam. It is now immediate, comparing the exponents of the two last expressions in Eq. (20), to get the optical theorem in this case:

\[
\sigma = \frac{4\pi}{k} Im f(0).
\]

and from it, using equations (19) and (8), to obtain the scattering cross section from a charged oscillating dipole:

\[
\sigma = 4\pi k \alpha'' = \frac{8\pi r_0^2}{3} \frac{\omega^4}{(\omega_0^2 - \omega^2)^2 + \gamma^2 \omega^2}.
\]

In Eq. (22), \(r_0 = e^2/mc^2\) is the classical electron radius, and we used the relation \(\gamma = 2e^2\omega^2/3mc^3\) for the radiation damping of Eq. (8). This important result, describing the Rayleigh scattering, incidentally explains, at an elementary level, the blue color of the sky we see when our line of sight forms a right angle with the white sunlight crossing the atmosphere, as well as its red color at sunrise and sunset.
In the opposite case of dense material, the relation between the atomic polarizability and the index of refraction is more complex and the formula of Lorentz-Lorenz is obtained solving Eq.(15) for the polarizability:

\[ \frac{n^2 - 1}{n^2 + 2} = \frac{4\pi}{3} \rho \alpha. \]  

(23)

III. NOTES ABOUT THE EXTINCTION THEOREM AND THE FIELDS GENERATED BY THE POLARIZATION CURRENTS

The previous results, in spite of the fact that they are well-known, are only seldom discussed in introductory electromagnetism and optics courses. This, notwithstanding the fact that some of them could easily be deduced from simple experimental observations.

In Fig. (1) an elementary experiment of optics is shown, that is commonly done to demonstrate Snell’s laws of reflection and refraction. A light beam is incident at an angle on a plane air-glass interface, and reflection and refraction beams are produced, all in the same plane of incidence. Using his exceptional physical intuition, Feynman interprets this sim-

FIG. 1. Experiment of a light beam striking obliquely an air-glass interface. Incident, reflected, and refracted beams are shown. The dotted line would be the direction of the incident wave in glass if it could go straight into it.
ple experimental situation in the light of the atomic model of matter to deduce, with the aid of the energy conservation law, the characteristics of the reflected beam.

He notices that a beam along the dotted line in fig.(1), which continues the incident beam straight into the dielectric, is not observed. It should, only if the atoms of the glass were frozen in their positions and could not oscillate under the driving action of the incoming beam, so that the beam can propagate along the dotted line into the interatomic vacuum with speed $c$. But the atoms do polarize and oscillate driven by the incoming primary field and by the secondary re-radiated fields by the multiple scattering atomic actions. The total induced polarization current must thus generate, in addition to the refracted beam, another vacuum beam that exactly cancels the incoming beam inside the dielectric. This is the way Feynman uses the extinction theorem (without ever naming it), just running the argument backwards: a vacuum beam inside is not observed, so, by superposition, there must be an internally generated vacuum beam that extinguishes it.

Equally interesting is to study the role of the polarization direction (perpendicular or parallel to the plane of incidence) of the incident beam in this experiment. As is well-known, these two electric field polarizations lead to different expressions for the reflection (and refraction) coefficients. Feynman approaches the problem of deducing the different contributions for the two cases according to the idea that only the fields radiated by the projections of the oscillating dipoles perpendicular to the propagation directions of both reflection and extinction fields are non-zero. He carries out the main calculation steps, also offering hints to arrive at the final answers that he writes down. A detailed derivation along these lines of thought, with some further interesting extensions, have recently been published\[11\].

Finally, there remains one more question to be noted about the experiment in fig.(1). The standard derivation for the reflection and transmission coefficients, for both perpendicular and parallel polarizations, is by matching boundary conditions at the air-glass interface for the macroscopic fields of Maxwell’s equations, and of course that works very well except that it does not clarify an important point. In fact, the reflected and transmitted fields appear, in that derivation, to only depend on what happens at the interface, a fact that seems to be also supported by examining the experimental situation in fig.(1). However, this is not what we understand with our microscopic calculation, which suggests instead a different physical interpretation. What generates all of the internal and external fields
(except the incident one) is the total polarization current that is set up into the dielectric material. If the material has a plane interface, as in fig. (1), then the fields radiated by all the atomic dipoles in V, coherently organise themselves in order to generate (in addition to the extinction field) fields that propagate along the refraction the reflected directions. This sounds more likely as a bulk effect rather than a surface effect.

Appendix A: Derivation of the results of (12) and (13).

The following standard vector identities involving the operator $\nabla$ hold:

$$\nabla \times (\vec{A} \times \vec{B}) = \vec{A}(\nabla \cdot \vec{B}) - \vec{B}(\nabla \cdot \vec{A}) + (\vec{B} \cdot \nabla)\vec{A} - (\vec{A} \cdot \nabla)\vec{B}$$

$$\nabla (\vec{A} \cdot \vec{B}) = (\vec{A} \cdot \nabla)\vec{B} + (\vec{B} \cdot \nabla)\vec{A} + \vec{A} \times (\nabla \times \vec{B}) + \vec{B} \times (\nabla \times \vec{A})$$

If $\vec{A}$ is a constant vector, then the terms that apply derivatives to $\vec{A}$ are zero, obtaining

$$\nabla \times (\vec{A} \times \vec{B}) = \vec{A}(\nabla \cdot \vec{B}) - (\vec{A} \cdot \nabla)\vec{B}$$

$$\nabla (\vec{A} \cdot \vec{B}) = (\vec{A} \cdot \nabla)\vec{B} + (\vec{B} \cdot \nabla)\vec{A} + \vec{A} \times (\nabla \times \vec{B}) + \vec{B} \times (\nabla \times \vec{A})$$

The complete expression for the Eq.(12) is thus:

$$\vec{E}_{rad,m}(\vec{r}_n) = -\nabla \times \left( \frac{\vec{p}_n \times \vec{r}_{nm}}{r_{nm}^3} \right) = \vec{v} \times \left( \nabla \times \frac{\vec{r}_{nm}}{r_{nm}^3} \right) - \vec{p}_n \left( \nabla \cdot \frac{\vec{r}_{nm}}{r_{nm}^3} \right)$$  \hspace{1cm} (A1)

The volume integral of Eq. (11) is thus composed by the three terms that appear in Eq.(A1), and it can be evaluated using the geometry shown in Fig. (2). The integral of the first term of (A1) is converted to a surface integral. The integral over the needle-shaped surface of the cylinder gives a negligible contribution, while the one on the surface $S_0$ results in the contribution reported in (13). This can be seen using $d\vec{a}_m = \hat{r}_{nm} r_{nm}^2 \sin \theta \, d\theta \, d\phi = -\hat{r}_{nm} r_{nm}^2 \, d\Omega_m$, and arbitrary choosing $\vec{p}_n = \hat{z} \hat{p}_n$. With these choices, we thus obtain

$$\int_{S_0-cyl} d^3 r_m \vec{E}_{rad,m}(\vec{r}_n) = \int_{S_0} d\vec{a}_m \left( \frac{\vec{p}_n \cdot \hat{r}_{nm}}{r_{nm}^3} \right) = -\vec{p}_n \int_{S_0} d\Omega_m \hat{r}_{nm} \cdot \hat{z} = -\frac{4\pi}{3} \vec{p}_n. \hspace{1cm} (A2)$$
FIG. 2. Geometry used to evaluate the integral of Eq. (11). On the left, the differential vectorial area $d\vec{a}_m$ is shown which as usual points to the outside of the closed surface $S_0$, centered at the position of the $n$-th atom. On the right, the polar coordinate system is shown, with the dipole moment $\vec{p}_n$ chosen to be in the $z$-direction. Note that the unit vector pointing to the outside is $\hat{r}_{mn} = -\hat{r}_{nm}$.

The second term of (A1) is easily seen to give a zero contribution to the integral:

$$\int_{S_0-cil} d^3r_m \vec{E}_{rad,m}(\vec{r}_n) = -\vec{p}_n \times \int_{S_0-cil} d^3r_m \left( \nabla_m \times \frac{\vec{r}_{nm}}{r_{nm}^3} \right) = -\vec{p}_n \times \int_{S_0-cil} d\vec{a}_m \times \frac{\vec{r}_{nm}}{r_{nm}^3} = 0 \quad (A3)$$

Finally, the third term of (A1) is not contributing since the origin is not included in the integration domain:

$$\int_{S_0-cil}^{(III)} d^3r_m \vec{E}_{rad,m}(\vec{r}_n) = \vec{p}_n \int_{S_0-cil} d^3r_m \left( \nabla_m \cdot \frac{\vec{r}_{nm}}{r_{nm}^3} \right) = \vec{p}_n \int_{S_0-cil} d^3r_m \delta(\vec{r}_{nm}) = 0 \quad (A4)$$

This demonstrates the result reported in Eq. (13).

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* giancarlo.reali@unipv.it

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