Microscopic derivation of the constitutive relations

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Abstract

Homogenization theory is used to deduce the macroscopic constitutive relations from the microscopic response function given by quantum-mechanical calculations.

1 Introduction

One can notice two trends in the investigation of the electromagnetic constitutive relations. The first trend, that could be called the formal approach, derives constitutive relations from general principles (thermodynamics [1], Lorentz covariance [2, 3], causality, time-invariance and continuity [4]). The second trend, called the constructive approach, derives constitutive relations from a microscopic description: constitutive relations are considered to be valid when a microscopic model is built, that exhibits the corresponding constitutive relations. The second approach is used implicitly by the theoreticians who calculate optical properties of solids (see Ref.[5] for a recent review).

The point that will be discussed in the present paper is the fact that the transition from a microscopic to a macroscopic constitutive relation is not simple. The usual random field approximation for the optical response of a quantum system gives the microscopic polarization field $\mathbf{p}(\mathbf{r})$ in terms of the microscopic electric field $\mathbf{e}(\mathbf{r})$,

$$p_i(\mathbf{r}) = \sum_j \int d\mathbf{r}' \chi_{ij}(\mathbf{r}, \mathbf{r}') e_j(\mathbf{r}'),$$

where the dielectric susceptibility $\chi(\mathbf{r}, \mathbf{r}')$ is the result of the quantum calculation. For crystals, the susceptibility has the periodicity $\chi(\mathbf{r}+\mathbf{R}, \mathbf{r}'+\mathbf{R}) = \chi(\mathbf{r}, \mathbf{r}')$ for every lattice vector $\mathbf{R}$.

In the optical range, the wavelength of the incident light is large compared to the crystal cell dimensions. Therefore, the optical properties of the crystal can be described correctly by using macroscopic fields $\mathbf{D}(\mathbf{r})$, $\mathbf{E}(\mathbf{r})$ and $\mathbf{P}(\mathbf{r})$. The macroscopic fields are obtained by averaging the microscopic fields over a crystal unit cell.

The problem is to find the macroscopic constitutive relation relating $\mathbf{P}(\mathbf{r})$ to $\mathbf{E}(\mathbf{r})$ from the microscopic ones. Homogenization theory is perfectly suited to this purpose, and will be adapted here to the case of a non-local susceptibility $\chi(\mathbf{r}, \mathbf{r}')$.

This paper, which summarizes Ref.[6], starts with an introduction to homogenization theory, then the microscopic Maxwell equations are homogenized to yield the macroscopic Maxwell equations and the constitutive relations.

2 The point of view of homogenization theory

Ever since the nineteenth century, physicists have homogenized microscopic systems by performing averages over distances very small compared to the macroscopic wavelength and very large compared to the atomic dimensions. In mathematics, homogenization is not considered as an averaging operation, but as a limit process. As an example, consider that a microscopic quantity can be represented by the periodic function $f(x) = \sin x + b$. To express the fact that the oscillation is very fast, mathematicians tried to give a meaning to the limit of $f(x/a)$ as $a \to 0$. In other words, what is the limit of a periodic function when its period becomes infinitely small ?

The contact with the physical point of view comes from the fact that the limit is the average of $f(x)$ over a period : $f(x/a) \to b$ as $a \to 0$. For a differential equation with rapidly oscillating coefficients, homogenization theory determines whether the solution has a limit when the period tends to zero, and to which equation the limit is a solution. This is exactly what we need to derive constitutive relations from a microscopic description of matter.

As a first step, we need to define a two-scale transform as follows. Take a three-dimensional periodic lattice with Brillouin zone BZ. Let C be the Wigner-Seitz cell of the lattice and $|C|$ its volume. The crystal sites will be denoted by $\mathbf{R}_s$, and the reciprocal lattice vectors by $\mathbf{K}$.

For a function f with Fourier transform $\tilde{f}(\mathbf{q})$ we define the two-scale transform of f as $\tilde{\mathsf{f}}(\mathbf{q},\rho) = \sum_{\mathbf{K}} \exp(i\mathbf{K}\cdot\rho)\tilde{f}(\mathbf{q}+\mathbf{K})$ for $\mathbf{q} \in BZ$ and $\tilde{\mathsf{f}}(\mathbf{q},\rho) = 0$ otherwise.

Since it is clear that $f(\mathbf{r}) = \mathsf{f}(\mathbf{r},\mathbf{r})$ (see however Ref.[7]), we have $f(\mathbf{r}) = \int_{BZ} d\mathbf{q} \exp(i\mathbf{q}\cdot\mathbf{r})\tilde{\mathsf{f}}(\mathbf{q},\mathbf{r})$.

From a microscopic function $f(\mathbf{r})$ we define a macroscopic function $F(\mathbf{r})$ by averaging $f(\mathbf{r})$ over a unit cell. More precisely

$$F(\mathbf{r}) \;=\; \langle f(\mathbf{r})\rangle = \int_{BZ} d\mathbf{q}\frac{1}{|C|}\int_C d\rho\, \tilde{\mathsf{f}}(\mathbf{q},\rho)\exp(i\mathbf{q}\cdot\mathbf{r}) = \int_{BZ} d\mathbf{q}\tilde{f}(\mathbf{q})\exp(i\mathbf{q}\cdot\mathbf{r}).$$

3 Homogenizing the Maxwell equations

To simplify the presentation, we consider a non magnetic sample and we neglect the effect of spin (see Ref.[8] for the full theory). Moreover, the electromagnetic charges, currents and fields have a time dependence $\exp(-i\omega t)$ which will be implicit for notational convenience.

The vacuum (rationalized SI) Maxwell equations describe the connection between the total fields and the induced charge and current densities:

$$\nabla\cdot\mathbf{e}(\mathbf{r}) = n(\mathbf{r})/\epsilon_0 \quad \nabla\times\mathbf{e}(\mathbf{r}) = i\omega\mathbf{b}(\mathbf{r}) \quad \nabla\cdot\mathbf{b}(\mathbf{r}) = 0 \quad \nabla\times\mathbf{b}(\mathbf{r}) + i\omega\epsilon_0\mu_0\mathbf{e}(\mathbf{r}) = \mu_0\mathbf{j}(\mathbf{r}),$$

where charge conservation implies $\nabla\cdot\mathbf{j}(\mathbf{r}) = i\omega n(\mathbf{r})$.

From the microscopic current $\mathbf{j}(\mathbf{r})$ it is customary, in dielectric theory, to define the polarization $\mathbf{p}(\mathbf{r}) = (i/\omega)\mathbf{j}(\mathbf{r})$ and the displacement field $\mathbf{d}(\mathbf{r}) = \epsilon_0\mathbf{e}(\mathbf{r}) + \mathbf{p}(\mathbf{r})$. In terms of those fields, the microscopic Maxwell equations become

$$\nabla\times\mathbf{e}(\mathbf{r}) = i\omega\mathbf{b}(\mathbf{r}) \quad \nabla\cdot\mathbf{b}(\mathbf{r}) = 0 \quad \nabla\cdot\mathbf{d}(\mathbf{r}) = 0 \quad \nabla\times\mathbf{b}(\mathbf{r}) = -i\omega\mu_0\mathbf{d}(\mathbf{r}). \tag{1}$$

Using the fact that the two-scale transform of $\nabla f(\mathbf{r})$ is $(i\mathbf{q}+\nabla_\rho)\tilde{\mathsf{f}}(\mathbf{q},\rho)$, we can make the two-scale transform of the Maxwell equations (1) to obtain:

$$\begin{aligned} i\mathbf{q}\times\mathbf{e}(\mathbf{q},\rho) + \nabla_\rho\times\mathbf{e}(\mathbf{q},\rho) &= i\omega\mathbf{b}(\mathbf{q},\rho) \qquad i\mathbf{q}\cdot\mathbf{b}(\mathbf{q},\rho) + \nabla_\rho\cdot\mathbf{b}(\mathbf{q},\rho) = 0 \\ i\mathbf{q}\cdot\mathbf{d}(\mathbf{q},\rho) + \nabla_\rho\cdot\mathbf{d}(\mathbf{q},\rho) &= 0 \qquad i\mathbf{q}\times\mathbf{b}(\mathbf{q},\rho) + \nabla_\rho\times\mathbf{b}(\mathbf{q},\rho) = -i\omega\mu_0\mathbf{d}(\mathbf{q},\rho). \end{aligned} \tag{2}$$

For notational convenience, we do not write the fields $\mathbf{e}(\mathbf{q},\rho)$, etc, with a tilde.

Consider a scattering problem where a plane electromagnetic wave is shined on the dielectric. In equations (2), the order of magnitude of $\mathbf{q}$ is $2\pi/\lambda$, where λ is the wavelength of the incident wave. The order of magnitude of the unit cell dimensions is $l = |C|^{1/3}$. Let $a = l/\lambda$, the order of magnitude of $\nabla_\rho \cdot \mathbf{e}(\mathbf{q},\rho)$ is $1/a$ times the order of magnitude of $\mathbf{q}\cdot\mathbf{e}(\mathbf{q},\rho)$. Now we expand all fields as sums of the type $\mathbf{e}(\mathbf{q},\rho) = \mathbf{e}^{(0)}(\mathbf{q},\rho) + a\mathbf{e}^{(1)}(\mathbf{q},\rho) + a^2\mathbf{e}^{(2)}(\mathbf{q},\rho) + \cdots$ where all terms of the expansion are periodic in ρ. Since a is small, we keep only the first term of the expansion to define the macroscopic fields as $\mathbf{E}(\mathbf{q}) = \langle \mathbf{e}^{(0)}(\mathbf{q},\rho)\rangle$, etc.

Introducing the expansions over a into Eq.(2) and gathering all terms of same power of a we obtain four equations for the a^{-1} term:

$$\nabla_\rho \times \mathbf{e}^{(0)}(\mathbf{q},\rho) = 0 \quad \nabla_\rho \cdot \mathbf{b}^{(0)}(\mathbf{q},\rho) = 0 \quad \nabla_\rho \cdot \mathbf{d}^{(0)}(\mathbf{q},\rho) = 0 \quad \nabla_\rho \times \mathbf{b}^{(0)}(\mathbf{q},\rho) = 0, \tag{3}$$

and four equations for the a^0 term:

$$\begin{aligned} i\mathbf{q}\times\mathbf{e}^{(0)}(\mathbf{q},\rho) + a\nabla_\rho\times\mathbf{e}^{(1)}(\mathbf{q},\rho) &= i\omega\mathbf{b}^{(0)}(\mathbf{q},\rho) \quad i\mathbf{q}\cdot\mathbf{b}^{(0)}(\mathbf{q},\rho) + a\nabla_\rho\cdot\mathbf{b}^{(1)}(\mathbf{q},\rho) = 0 \qquad (4)\\ i\mathbf{q}\cdot\mathbf{d}^{(0)}(\mathbf{q},\rho) + a\nabla_\rho\cdot\mathbf{d}^{(1)}(\mathbf{q},\rho) &= 0 \quad i\mathbf{q}\times\mathbf{b}^{(0)}(\mathbf{q},\rho) + a\nabla_\rho\times\mathbf{b}^{(1)}(\mathbf{q},\rho) = -i\omega\mu_0\mathbf{d}^{(0)}(\mathbf{q},\rho), \end{aligned}$$

(recall that $a\nabla_\rho$ has the same order of magnitude as $\mathbf{q}$). These two sets of equations are sufficient to determine the macroscopic Maxwell equations.

To obtain the macroscopic Maxwell equations, we must eliminate the terms of order one, e.g. $\mathbf{b}^{(1)}(\mathbf{q},\rho)$. This is achieved by transforming the cell average of the divergence into an integral over the surface ∂C of the unit cell: $\langle \nabla_\rho \cdot \mathbf{b}^{(1)}(\mathbf{q},\rho)\rangle = 1/|C| \int_{\partial C} \hat{n}\cdot\mathbf{b}^{(1)}(\mathbf{q},\rho)dS$. On opposite sides of cell C, the outgoing normal $\hat{n}$ is reversed while $\mathbf{b}^{(1)}(\mathbf{q},\rho)$ is equal (by periodicity). Therefore, the surface integral is zero and $\langle \nabla_\rho \cdot \mathbf{b}^{(1)}(\mathbf{q},\rho)\rangle = 0$. A similar reasoning leads to $\langle \nabla_\rho \times \mathbf{b}^{(1)}(\mathbf{q},\rho)\rangle = 0$.

Thus, averaging Eq.(4) gives $0 = i\mathbf{q}\cdot\langle \mathbf{b}^{(0)}(\mathbf{q},\rho)\rangle + a\langle \nabla_\rho \cdot \mathbf{b}^{(1)}(\mathbf{q},\rho)\rangle = i\mathbf{q}\cdot\mathbf{B}(\mathbf{q})$, because the macroscopic field $\mathbf{B}(\mathbf{q})$ was defined as the average of $\mathbf{b}^{(0)}(\mathbf{q},\rho)$. Carrying out similar calculations for the other microscopic Maxwell equations we obtain the usual macroscopic Maxwell equations in momentum space.

4 Macroscopic constitutive relations

To derive the macroscopic constitutive relations, we start from the two-scale transform of the microscopic constitutive relations: $\mathbf{d}(\mathbf{q},\rho) = \epsilon_0\mathbf{e}(\mathbf{q},\rho) + \tilde{\chi}(\rho,\rho';\mathbf{q})\cdot\mathbf{e}(\mathbf{q},\rho)$.

From Eqs.(3), we see that there is a periodic potential (with zero average) $\phi(\mathbf{q},\rho)$ such that $\mathbf{e}^{(0)}(\mathbf{q},\rho) = \mathbf{E}(\mathbf{q}) - \nabla_\rho\phi(\mathbf{q},\rho)$. Moreover, $\nabla_\rho \cdot \mathbf{d}^{(0)}(\mathbf{q},\rho) = 0$ enables us to write the equation

$$\epsilon_0\Delta_\rho\phi(\mathbf{q},\rho) + \sum_{ij}\langle \partial_{\rho_i}\tilde{\chi}_{ij}(\rho,\rho';\mathbf{q})\partial_{\rho'_j}\phi(\mathbf{q},\rho')\rangle_{\rho'} = \sum_{ij}\langle \partial_{\rho_i}\tilde{\chi}_{ij}(\rho,\rho';\mathbf{q})\rangle_{\rho'}E_j(\mathbf{q}). \tag{5}$$

which determines $\phi(\mathbf{q},\rho)$ uniquely.

Let the three functions $A_k(\mathbf{q},\rho)$ ($k = x,y,z$) be the solutions of Eq.(5) for an electric field $\mathbf{E}$ equal to a unit vector in the direction k. Then, for a general electric field $\mathbf{E}(\mathbf{q})$, the potential is $\phi(\mathbf{q},\rho) = \mathbf{A}(\mathbf{q},\rho)\cdot\mathbf{E}(\mathbf{q})$.

If we use this expression for $\phi(\mathbf{q},\rho)$ and average over the unit cell to obtain macroscopic fields, we find the macroscopic constitutive relation $D_i(\mathbf{q}) = \sum_j \epsilon_{ij}(\mathbf{q})E_j(\mathbf{q})$ where the macroscopic dielectric constant $\epsilon_{ij}(\mathbf{q})$ is given by

$$\epsilon_{ij}(\mathbf{q}) \quad = \quad \epsilon_0\delta_{ij} + \sum_k \langle \tilde{\chi}_{ik}(\rho,\rho';\mathbf{q})[\delta_{kj} - \partial_{\rho'_k}A_j(\mathbf{q},\rho')]\rangle_{\rho\rho'}.$$

In the real space, this gives us $D_i(\mathbf{r}) = \sum_j \int d\mathbf{r}'\bar{\epsilon}_{ij}(\mathbf{r}-\mathbf{r}')E_j(\mathbf{r}')$. This equation is typical of a homogeneous (but generally anisotropic) non-local medium.

5 Conclusion

For the standard theory of dielectrics, we have shown that the macroscopic constitutive relations have the same form as the microscopic ones, although they are not given by the average of the microscopic constitutive relations.

In general, homogenization theory has shown that the macroscopic equation can be strongly different from the microscopic one. For instance, instantaneous microscopic equations can turn into equations with memory, local equations can develop non-local macroscopic terms, mixtures of optically inactive materials can become optically active [9].

The macroscopic and the microscopic constitutive relations are similar because the polarization of the medium was entirely described by $\mathbf{p}(\mathbf{r})$. This is natural in the minimal coupling description of the interaction between light and matter. If the Power-Zienau-Woolley transform is applied (or the Poincaré gauge is used), then the polarization $\mathbf{p}(\mathbf{r})$ and the magnetization $\mathbf{m}(\mathbf{r})$ appear naturally in the microscopic Maxwell equations and the macroscopic constitutive relations are modified.

This point is related to the fact that the Maxwell equations do not define $\mathbf{p}(\mathbf{r})$ and $\mathbf{m}(\mathbf{r})$ uniquely. For any vector field $\mathbf{G}(\mathbf{r})$ and scalar field $g(\mathbf{r})$, the polarization and magnetization $\mathbf{p}' = \mathbf{p} + \nabla \times \mathbf{G}$ and $\mathbf{m}' = \mathbf{m} + \nabla g - \partial \mathbf{G}/\partial t$ satisfy the same Maxwell equations [10]. The only changes being in the boundary conditions (the surface charge and currents are different) [11].

A way out of this ambiguity would be to determine the constitutive relation that lead to the simplest boundary conditions. In principle, homogenization enables us to calculate the boundary conditions through a boundary layer at the surface. This work is in progress.

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