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Quantization and Microscopic Background of Macroscopic Dielectrics

Abstract

A microscopic linear response expression for the electric susceptibility of a finite dielectric is presented. Its form justifies the use of the experimentally accessible macroscopic susceptibility in the quantisation of phenomenological absorptive dielectrics.

1 Background

Quantization of macroscopic dielectrics is required for a proper description of processes such as the radiative decay of atoms embedded in absorptive photonic crystals, transition and Čerenkov X-ray radiation by fast electrons moving through absorptive dielectric layers, the Casimir force between absorptive media and quantum friction. In particular the atomic decay and X-ray cases have important technological implications.

The quantization of conservative (non-absorptive) media, characterized by the real frequency-independent electric permeability $\varepsilon(\mathbf{x})$, has a long history but a corresponding approach for the absorptive case, involving a complex, frequency-dependent $\varepsilon(\mathbf{x},\omega)$, has only become available quite recently and is at the moment still confined to the linear case. Two general approaches exist:

- The addition of a Langevin noise current [1, 2, 3, 4], describing the absorption, to the free quantized free field equations.
- The introduction of auxiliary fields in the classical absorptive case [5], restoring energy conservation and allowing a canonical formalism and its quantisation.

In these setups the only input required is $\varepsilon(\mathbf{x},\omega)$, which can be obtained from experiment. Another line of attack starts from a microscopic quantum model for the material system [6, 7, 8] So far its scope is limited to spatially homogeneous situations and it uses a specific model for the material system. This situation leads to the following questions:

- Are the two macroscopic approaches equivalent?
- Can a microscopic justification be given that goes beyond [6, 7, 8]?

Both questions have an affirmative answer. It is easy to show that the auxiliary field model immediately leads to a Langevin current with the correct properties. In fact the converse route can be followed as well. The second problem is more intriguing. An obvious starting

point would be some linear (and higher order) response approach, involving a material system interacting with the free electromagnetic field, followed by the derivation of an effective set of equations for the electromagnetic subsystem. Its feasibility strongly depends on the actual material system. The first choice to be made is that between an infinite and finite system. In actual situations one is dealing with a finite piece of material, which is often so large that its spectrum approaches the simpler band structure of the infinite system in the crystalline case, suggesting the use of an infinite system to exploit this feature. However, there is an important drawback. In order to study the dielectric properties of the system an electric field is generated outside the material and its effect is studied with outside detectors. This gives serious problems in the infinite case. But even in the finite case a linear response expression may be meaningless (consider a single moving charge). Assuming our piece of material to be initially in a bound state (i.e., we look upon it as a large molecule) and the initial field state that of a wavepacket moving towards the material, we have a realistic situation which is amenable to a scattering theoretical description. The idea is then to derive a set of Maxwell's equations for the field subsystem. This set is not unique but will in general depend on the observed process. Elastic scattering of the field wavepacket from the material can be handled by linear response theory, the coupling constant between the subsystems being the small fine structure constant α. Non-linear processes, such as harmonic generation, require a higher order response approach.

2 The system

We consider a finite material system, made up from spinless Schrödinger particles interacting through Coulomb forces, which is coupled to the transverse quantized Maxwell field. Its Hamiltonian is

$$H = H_m + H_f + H_{int} = H_0 + H_{int} = H_m + H_f - \int d\mathbf{x} \boldsymbol{J}^{\perp}(\mathbf{x}) \cdot \boldsymbol{A}(\mathbf{x}), \tag{1}$$

$$H_m = \sum_{j=1}^{N} \frac{\mathbf{p}_j^2}{2m_j} + \sum_{i>j=1}^{N} V_{ij}(\mathbf{x}_i - \mathbf{x}_j), \quad H_f = \sum_{\alpha} \int d\mathbf{k} k a^*(\mathbf{u}_{\mathbf{k}\alpha}) a(\mathbf{u}_{\mathbf{k}\alpha}), \tag{2}$$

$$\boldsymbol{A}(\mathbf{x}) = \sum_{\alpha} \int d\mathbf{k} (2k)^{-1/2} \{a^*(\mathbf{u}_{\mathbf{k}\alpha}) \tilde{\mathbf{u}}_{\mathbf{k}\alpha}(\mathbf{x}) + a(\mathbf{u}_{\mathbf{k}\alpha}) \mathbf{u}_{\mathbf{k}\alpha}(\mathbf{x})\} \tag{3}$$

where H_m is the matter Hamiltonian, consisting of a kinetic energy term and a sum of Coulomb interactions, H_f the free field Hamiltonian, $\boldsymbol{A}(\mathbf{x})$ the vector potential and $\boldsymbol{J}^{\perp}(\mathbf{x})$ the transverse part of the current operator

$$\boldsymbol{J}(\mathbf{x}) = \sum_j \frac{e_j}{2m_j} \{\mathbf{p}_j \delta(\mathbf{x} - \mathbf{x}_j) + \delta(\mathbf{x} - \mathbf{x}_j) \mathbf{p}_j\} - \sum_j \frac{e_j^2}{m_j} \delta(\mathbf{x} - \mathbf{x}_j) \, \boldsymbol{A}(\mathbf{x}_j) = \boldsymbol{J}_a(\mathbf{x}) + \boldsymbol{J}_b(\mathbf{x}). \tag{4}$$

Here the a^*'s and a's are creation and annihilation operators and the $\mathbf{u}_{\mathbf{k}\alpha}$ are the free transverse free field modes. Initially, as $t \to -\infty$ the system approaches the freely evolving product state

$$\rho_{in} = \rho_m \otimes \rho_f. \tag{5}$$

Here ρ_m is the density operator describing the initial matter state, which we assume to commute with H_m, $[\rho_m, H_m] = 0$, whereas ρ_f is the density operator for the initial field wavepacket. It can be constructed in such a way that it does not reach the material target untill some finite time t which we set equal to zero. This cannot strictly be true, since eigenvectors of Schrödinger operators have infinite tails. The latter decay rapidly over a distance in the order of Ångstrøms and it makes sense to set the system density operator at $t = 0$ equal to ρ_{in},

$$\rho(0) \approx \rho_{in}. \tag{6}$$

This description can be improved, using wave operators to relate the initial situation to that at $t = 0$. The time evolution of an observable in the Heisenberg picture is given by $(LX = [H, X])$

$$X(t) = \exp[iHt]X \exp[-iHt] = \exp[Lt\}X = U(t)X \tag{7}$$

and for the field operators this leads to

$$\begin{aligned} \partial_t \boldsymbol{E}(\mathbf{x}, t) &= \partial_{\mathbf{x}} \times \boldsymbol{B}(\mathbf{x}, t) - \boldsymbol{J}^{\perp}(\mathbf{x}, t), & (8) \\ \partial_t \boldsymbol{B}(\mathbf{x}, t) &= -\partial_{\mathbf{x}} \times \boldsymbol{E}(\mathbf{x}, t). & (9) \end{aligned}$$

We then obtain effective equations for the field operators by taking the (partial) trace over the initial matter density operator, i.e., denoting $\langle X \rangle = \mathrm{tr}_m \rho_m X$, we have

$$\begin{aligned} \partial_t \langle \boldsymbol{E} \rangle(\mathbf{x}, t) &= \partial_{\mathbf{x}} \times \langle \boldsymbol{B} \rangle(\mathbf{x}, t) - \langle \boldsymbol{J} \rangle^{\perp}(\mathbf{x}, t), & (10) \\ \partial_t \langle \boldsymbol{B} \rangle(\mathbf{x}, t) &= -\partial_{\mathbf{x}} \times \langle \boldsymbol{E} \rangle(\mathbf{x}, t). & (11) \end{aligned}$$

or

$$-\partial_t^2 \langle \boldsymbol{A} \rangle(\mathbf{x}, t) = -\partial_{\mathbf{x}}^2 \langle \boldsymbol{A} \rangle(\mathbf{x}, t) - \langle \boldsymbol{J} \rangle^{\perp}(\mathbf{x}, t). \tag{12}$$

3 Linearisation

A expression for $\boldsymbol{J}^{\perp}(\mathbf{x}, t)$, linear in the fields, can be obtained by making a Dyson series expansion of $U(t)$ with $L_{int}X = [H_{int}, X]$ as the perturbation and retaining the first two terms. Thus, in terms of $U_0(t)$ $(U_\gamma(t) = \exp[iL_\gamma t]$, $L_\gamma X = [H_\gamma, X])$,

$$U(t) = U_0(t) + i \int_0^t ds U_0(t-s) L_{int} U_0(s) + \mathcal{O}(L_{int}^2). \tag{13}$$

However, note that L_{int} contains linear and quadratic contributions in $\boldsymbol{A}$. Expanded in increasing orders in $\boldsymbol{A}$, we have

$$\boldsymbol{J}^{\perp}(\mathbf{x}, t) = \sum_{n=0}^{\infty} \boldsymbol{J}_n^{\perp}(\mathbf{x}, t), \tag{14}$$

where

$$\boldsymbol{J}_0^{\perp}(\mathbf{x},t) = U_0(t)\boldsymbol{J}_a^{\perp}(\mathbf{x}) = U_m(t)\boldsymbol{J}_a^{\perp}(\mathbf{x}), \tag{15}$$

$$\boldsymbol{J}_1^{\perp}(\mathbf{x},t) = U_0(t)\boldsymbol{J}_b^{\perp}(\mathbf{x}) - i\int_0^t ds \int d\mathbf{y} U_0(t-s)[\boldsymbol{J}_a^{\perp}(\mathbf{y})\cdot\ \boldsymbol{A}(\mathbf{y}), U_0(s)\boldsymbol{J}_a^{\perp}(\mathbf{x})], \tag{16}$$

etc. We now average over ρ_m, leading to $\langle \boldsymbol{J}_0^{\perp}(\mathbf{x},t)\rangle = \langle \boldsymbol{J}_a^{\perp}(\mathbf{x})\rangle$, which may or may not vanish, depending on the symmetry properties of ρ_m. Next, after some rewriting,

$$\langle \boldsymbol{J}_1^{\perp}(\mathbf{x},t)\rangle = \langle U_f(t)\boldsymbol{J}_b^{\perp}(\mathbf{x})\rangle - i\int_0^t ds \int d\mathbf{y}\langle[\boldsymbol{J}^{\perp}(\mathbf{y})\cdot\ U_f(t-s)\ \boldsymbol{A}(\mathbf{y}), U_m(s)\boldsymbol{J}_a^{\perp}(\mathbf{x})]\rangle, \tag{17}$$

where the second term has the structure of an autocorrelation expression. Here the right hand side features the freely evolving $U_f(u)\boldsymbol{A}(\mathbf{y})$ and our final step is to keep these first two terms in (12) in a self-consistent approximation where we replace $U_f(u)\boldsymbol{A}$ with $\langle \boldsymbol{A}\rangle(u)$, i.e. the same object as featured on the left hand side. Now it is straightforward to identify the quantity corresponding to the phenomenological electric susceptibility $\boldsymbol{\chi}$, relating the polarisation $\boldsymbol{P}$ to the electric field

$$\boldsymbol{P}(\mathbf{x},t) = \int_0^t ds \int d\mathbf{y}\boldsymbol{\chi}\ (\mathbf{x},\mathbf{y},t-s)\cdot\boldsymbol{E}(\mathbf{y},s), \tag{18}$$

which in general is a second rank tensor and a kernel in coordinate space as well. This is most easily done by switching to Laplace transforms,

$$\hat{f}(z) = \int_0^{\infty} dt \exp[izt] f(t), \quad \mathrm{Im} z > 0, \tag{19}$$

so

$$\widehat{\boldsymbol{E}}(z) = -\boldsymbol{E}(0) - iz\,\widehat{\boldsymbol{A}}(z), \tag{20}$$

which relation can be used to express the current in terms of $\widehat{\boldsymbol{E}}(z)$. Apparently the above straightforward procedure in this general form has not been considered in the literature, although it has recently been applied to a description of collective excitations [9].

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