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A single susceptibility scheme of macroscopic Maxwell equations: beyond the 'E, D, B, H' approach

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Abstract

A new single susceptibility scheme of macroscopic Maxwell equations has been derived by applying a long wavelength approximation to the general form of microscopic response represented by the simultaneous integral equations of induced current density and vector potential. The result turns out to be more general than the conventional '$\boldsymbol{E}$, $\boldsymbol{D}$, $\boldsymbol{B}$, $\boldsymbol{H}$' scheme. The new (single) susceptibility contains the contributions of electric and magnetic polarizations together with their mutual interference effect in its different order terms of wavevector $\boldsymbol{k}$. The conventional description via ϵ and μ can be reproduced only in the absence of chiral symmetry, under the additional condition that magnetic susceptibility defined with respect, not to $\boldsymbol{H}$, but to $\boldsymbol{B}$ should be used. In the presence of chiral symmetry, the phenomenological Drude–Born–Fedorov constitutive equations cannot be justified by this microscopic approach.

1. Introduction

Maxwell equations (M-eqs), one of the most fundamental theoretical schemes of physics, have two forms, i.e. micro- and macroscopic ones. The latter is an approximate scheme of the former appropriate for macroscopic matter systems. Historically, however, the former was first proposed when there was neither quantum mechanics nor relativity theory, so that it was a phenomenology. At the beginning of the 20th century, the microscopic form based on the particle picture of matter was proposed [1], and this, combined with quantum mechanics and relativity, led to quantum electrodynamics (QED), which is one of the most accurate theories of physics [2, 3].

There have been various attempts to derive the macroscopic M-eqs from the microscopic M-eqs based on the particle picture of matter [4]. Thereby, the standard way of thinking is as follows. The first step is to admit that the macroscopic average of the charge and current densities in the microscopic M-eqs can be expressed in terms of electric ($\boldsymbol{P}$) and magnetic polarizations ($\boldsymbol{M}$). Then, one separates (a) charge density ρ into true (ρ_{t}) and polarization charge density (ρ_{p}), and (b) the transverse component of current density into the contributions of electric and magnetic polarizations, $\partial \boldsymbol{P}/\partial t$ and $c\nabla \times \boldsymbol{M}$, respectively. By using the new variables $\boldsymbol{P}$, $\boldsymbol{M}$, $\boldsymbol{D} = \boldsymbol{E} + 4\pi \boldsymbol{P}$, and $\boldsymbol{H} = \boldsymbol{B} - 4\pi \boldsymbol{M}$, one can rewrite the microscopic M-eqs into the macroscopic M-eqs. In the case of linear response, one introduces (macroscopic) electric and magnetic susceptibilities, χ_{e} and χ_{m}, respectively, via $\boldsymbol{P} = \chi_{\mathrm{e}}\boldsymbol{E}$ and $\boldsymbol{M} = \chi_{\mathrm{m}}\boldsymbol{H}$, which define dielectric constant and magnetic permeability as $\epsilon = 1 + 4\pi\chi_{\mathrm{e}}$, $\mu = 1 + 4\pi\chi_{\mathrm{m}}$.

In terms of symmetry argument and quantum mechanical calculation, one can argue about the frequency and wavevector dependence of susceptibilities, so that the macroscopic M-eqs at this level can be regarded as semiquantitative rather than purely phenomenological theory. These types of macroscopic M-eqs in terms of the field variables '$\boldsymbol{E}$, $\boldsymbol{D}$, $\boldsymbol{B}$, $\boldsymbol{H}$' have been successfully used for studies of macroscopic electromagnetic (EM) response in a vast area, and played an indispensable role in the great development of modern physics and technology in the 20th century.

However, in spite of the great success, the macroscopic M-eqs seem to be incomplete in the rigorous sense of uniqueness and consistency, because (i) the unique way of separation (a) and (b) mentioned above is not known, and (ii) the question why the number of susceptibilities is different between the micro- and macroscopic M-eqs is not answered clearly. (In the microscopic M-eqs we need only one susceptibility tensor between induced current density $\boldsymbol{J}$ and vector potential $\boldsymbol{A}$, while we need two, χ_{e} and χ_{m}, in the macroscopic M-eqs [5].) In addition to these problems, there are some other questions, (iii) why the usual treatment of spin resonance [6] and orbital magnetic dipole transition [7] lead to different wavevector

dependence of μ, i.e. $O(k^0)$ for the former, and $O(k^2)$ for the latter, and (iv) why the conventional dispersion equation $(ck/\omega)^2 = \epsilon\mu$ have the contributions of electric and magnetic dipole transitions as a product on the rhs of the equation. Since, in low symmetry, there is no distinction between these two types of transitions, one should generally expect their contributions as a sum rather than a product.

There are proposals of single susceptibility scheme of macroscopic M-eqs by Agranovich *et al* [8] and Il'inskii and Keldysh [9]. Though their motivation is similar to (ii) mentioned above, their results do not answer all the questions mentioned above, and the relation with the conventional scheme is not clarified. (More discussion is given in section 3.2.) The purpose of this paper is to give a new single susceptibility scheme, which provides answers to all the questions (i)–(iv) and clarifies the relationship with the conventional '$\boldsymbol{E}, \boldsymbol{D}, \boldsymbol{B}, \boldsymbol{H}$' scheme.

In order to make a more complete form of single susceptibility theory, we take a new approach to derive the macro- from the microscopic M-eqs, i.e. we apply the long wavelength approximation (LWA) to the microscopic EM response of matter. This method relies only on the well-defined Lagrangian of matter and EM field and standard quantum mechanics, which allows mathematically clear-cut treatment, without using empirical knowledge and/or model-dependent concepts. It turns out that the new scheme is more general than the '$\boldsymbol{E}, \boldsymbol{D}, \boldsymbol{B}, \boldsymbol{H}$' scheme. In particular, it provides the first-principles macroscopic susceptibility, including the case of chiral symmetry, where electric and magnetic dipole transitions are mixed with one another.

The macroscopic M-eqs are still a main tool for the studies of photonic crystals [10], near-field optics [11], left-handed or metamaterials [12, 13], etc, it is desirable to establish a single susceptibility scheme which allows the first-principles treatment of coexisting electric and magnetic polarizations. It would also contribute to teaching M-eqs through a mathematically clear description of the hierarchical structure of the microscopic response theory and the macroscopic M-eqs.

2. Formulation

2.1. Microscopic response theory

As a reliable basis of the new macroscopic M-eqs, we first describe the microscopic response theory within the semi-classical regime. This consists of the microscopic M-eqs and the microscopic constitutive equation, and the response is given as the solution of simultaneous integral equations of vector potential and current density. Though this part of the theory is well established, we describe it in detail below, since its generality is the guarantee of the soundness of the new macroscopic M-eqs.

We start with the general Lagrangian of charged particles and EM field

$$L = \sum_\ell \left\{ \frac{1}{2} m_\ell v_\ell^2 - e_\ell \phi(\boldsymbol{r}_\ell) + \frac{e_\ell}{c} \boldsymbol{v}_\ell \cdot \boldsymbol{A}(\boldsymbol{r}_\ell) \right\} + \int \mathrm{d}\boldsymbol{r} \frac{1}{8\pi} \left\{ \left(\frac{1}{c} \frac{\partial \boldsymbol{A}}{\partial t} + \nabla\phi \right)^2 - (\nabla \times \boldsymbol{A})^2 \right\}, \quad (1)$$

where $\boldsymbol{A}$ and ϕ are vector and scalar potentials, e_ℓ, $\boldsymbol{r}_\ell$, and $\boldsymbol{v}_\ell$ the charge, coordinate, and velocity of the ℓth particle. The least action principle of this Lagrangian provides 'microscopic M-eqs' and the 'Newton equation of each particle under Lorentz force', so that this is a sufficiently reliable starting point. The Hamiltonian for the particles in an EM field is derived from this Lagrangian, in Coulomb gage ($\nabla \cdot \boldsymbol{A} = 0$), as

$$H_\mathrm{M} = \sum_\ell \frac{1}{2m_\ell} \left[\boldsymbol{p}_\ell - \frac{e_\ell}{c} \boldsymbol{A}(\boldsymbol{r}_\ell) \right]^2 + \frac{1}{2} \sum \sum_{\ell \neq \ell'} \frac{e_\ell e_{\ell'}}{|\boldsymbol{r}_\ell - \boldsymbol{r}_{\ell'}|}. \quad (2)$$

The Coulomb potential term is the sum of the scalar potential related terms (the integrals of $e\phi$ and $(1/c)(\nabla\phi)^2$). Since this term is usually regarded as a part of the matter Hamiltonian, the EM field interacting with the matter is represented by the transverse (T) field, $\boldsymbol{A}$, alone. If necessary, we can add relativistic correction terms, such as spin–orbit interaction, spin Zeeman interaction, etc to make the Hamiltonian general enough for the problems of materials science. The EM response of such materials can be treated by simply adding the Hamiltonian of the free EM field. It becomes a semi-classical EM response theory by quantizing the particle variables, and (non-relativistic) QED by further quantizing the EM field, so that this Lagrangian can be a fully general starting point of EM response theory within the non-relativistic regime.

The fundamental equations of semi-classical EM response are a set of M-eqs and constitutive equation(s). The latter is the relation between induced polarization(s) and source EM field in terms of susceptibility. This set of equations gives the self-consistent solution of EM field and induced polarizations, especially of their transverse components, which leads to the coupled eigenmodes, such as polaritons. To calculate the induced polarization as a linear response, we use the $\boldsymbol{A}$-linear term of (2), $H_\mathrm{lin} = (-1/c) \int \mathrm{d}\boldsymbol{r}\, \boldsymbol{J} \cdot \boldsymbol{A}$, where $\boldsymbol{J}$ is the current density operator $\boldsymbol{J}(\boldsymbol{r}) = \sum_\ell e_\ell \boldsymbol{v}_\ell \delta(\boldsymbol{r} - \boldsymbol{r}_\ell)$. In the presence of spin magnetization $\boldsymbol{M}_\mathrm{spin}(\boldsymbol{r}) = \sum_\ell \beta_\ell \boldsymbol{s}_\ell \delta(\boldsymbol{r} - \boldsymbol{r}_\ell)$, where $\beta_\ell \boldsymbol{s}_\ell$ is the spin magnetization of the ℓth particle with spin $\boldsymbol{s}_\ell$, we need to consider also its interaction with EM field, i.e. the spin Zeeman interaction $H_\mathrm{sZ} = -\int \mathrm{d}\boldsymbol{r} \boldsymbol{M}_\mathrm{spin} \cdot \boldsymbol{B} = (-1/c) \int \mathrm{d}\boldsymbol{r} \boldsymbol{J}_\mathrm{spin} \cdot \boldsymbol{A}$, where the spin induced current density is defined as $\boldsymbol{J}_\mathrm{spin}(\boldsymbol{r}) = c\nabla \times \boldsymbol{M}_\mathrm{spin}(\boldsymbol{r})$. Thus we can take the sum

$$H_\mathrm{int} = -\frac{1}{c} \int \mathrm{d}\boldsymbol{r}\, \boldsymbol{I}(\boldsymbol{r}) \cdot \boldsymbol{A}(\boldsymbol{r}, t), \qquad (\boldsymbol{I} = \boldsymbol{J} + \boldsymbol{J}_\mathrm{spin}) \quad (3)$$

as the generalized interaction Hamiltonian containing both orbital and spin current densities. Correspondingly, the matter Hamiltonian H_0 ($\boldsymbol{A} = 0$ term of H_M) should contain the remaining terms of relativistic correction. In this way, we can prepare the matter Hamiltonian and matter–EM field interaction H_int model-independently.

Induced polarization is calculated via standard time-dependent perturbation theory, found in various textbooks, e.g. [14–16], so that we only give an outline. The microscopic constitutive equation is obtained by calculating the expectation value of the current density operator with respect to the matter wavefunction at time t, $\langle \Psi(t) | \boldsymbol{I}(\boldsymbol{r}) | \Psi(t) \rangle$. The time

evolution of Ψ is governed by the Schrödinger equation $\mathrm{i}\hbar\partial\Psi/\partial t = (H_0 + H_{\rm int})\Psi$. In the interaction representation $\Psi = \exp(-\mathrm{i}H_0\tau/\hbar)\tilde{\Psi}$, the Schrödinger equation takes the form $\mathrm{i}\hbar\partial\tilde{\Psi}/\partial t = H'(t)\tilde{\Psi}$, where

$$H'(\tau) = \exp(\mathrm{i}H_0\tau/\hbar)H_{\rm int}\exp(-\mathrm{i}H_0\tau/\hbar). \tag{4}$$

For linear response we only need the lowest-order iterative solution

$$\tilde{\Psi}(t) = \tilde{\Psi}(-\infty) - \frac{\mathrm{i}}{\hbar}\int_{-\infty}^{t} \mathrm{d}\tau\, H'(\tau)\tilde{\Psi}(-\infty) + \cdots, \tag{5}$$

where we assume that the interaction $H_{\rm int}$ is adiabatically switched on in the remote past to the matter state in the ground state $|0\rangle$ of the Hamiltonian H_0, i.e. $\tilde{\Psi}(-\infty) = |0\rangle$.

The $\boldsymbol{A}$ linear terms of the expectation value $\langle\Psi(t)|\boldsymbol{I}(\boldsymbol{r})|\Psi(t)\rangle$ originate from those of $\Psi(t)$ and of the operator $\boldsymbol{I}$. We separate the $\boldsymbol{A}$ dependent part of $\boldsymbol{I}$ (arising from the velocity term $\boldsymbol{v} = (\boldsymbol{p} - e\boldsymbol{A}/c)/m$) as

$$\boldsymbol{I}(\boldsymbol{r}) = \hat{\boldsymbol{I}}(\boldsymbol{r}) - \frac{1}{c}\hat{N}(\boldsymbol{r})\boldsymbol{A}(\boldsymbol{r}, t) \tag{6}$$

where $\hat{N}(\boldsymbol{r}) = \sum_\ell (e_\ell^2/m_\ell)\delta(\boldsymbol{r} - \boldsymbol{r}_\ell)$. In terms of these notations, we can evaluate the $\boldsymbol{A}$ linear part of the induced current density (with frequency ω) as

$$\boldsymbol{I}(\boldsymbol{r},\omega) = -\frac{1}{c}\langle 0|\hat{N}(\boldsymbol{r})|0\rangle\boldsymbol{A}(\boldsymbol{r},\omega) + \frac{1}{c}\sum_\nu [g_\nu(\omega)\hat{\boldsymbol{I}}_{0\nu}(\boldsymbol{r})F_{\nu 0}(\omega) + h_\nu(\omega)\hat{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r})F_{0\nu}(\omega)], \tag{7}$$

where $\hat{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{r}) = \langle\mu|\hat{\boldsymbol{I}}(\boldsymbol{r})|\nu\rangle$ with $H_0|\mu\rangle = E_\mu|\mu\rangle$, etc,

$$F_{\mu\nu}(\omega) = \int \mathrm{d}\boldsymbol{r}\, \hat{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{r})\cdot\boldsymbol{A}(\boldsymbol{r},\omega) \qquad (\mu = 0,\ \text{or}\ \nu = 0), \tag{8}$$

and

$$g_\nu(\omega) = \frac{1}{E_{\nu 0} - \hbar\omega - i0^+}, \qquad h_\nu(\omega) = \frac{1}{E_{\nu 0} + \hbar\omega + i0^+}, \tag{9}$$

$E_{\nu 0} = E_\nu - E_0$. The first term of (7) mostly reflects the ground state charge density of electrons, which, in the LWA, leads to $-(e^2 n_0/mc)\boldsymbol{A}$, where n_0, m, e are the density, charge, and mass of electrons, respectively. This contributes to the macroscopic susceptibility as an additional constant. This is one way of treating this term, but there is another way, i.e. to renormalize it into the second term of (7) as

$$\boldsymbol{I}(\boldsymbol{r},\omega) = \frac{1}{c}\sum_\nu [\bar{g}_\nu(\omega)\hat{\boldsymbol{I}}_{0\nu}(\boldsymbol{r})F_{\nu 0}(\omega) + \bar{h}_\nu(\omega)\hat{\boldsymbol{I}}_{\nu 0}(\boldsymbol{r})F_{0\nu}(\omega)], \tag{10}$$

where

$$\bar{g}_\nu(\omega) = g_\nu(\omega) - \frac{1}{E_{\nu 0}}, \qquad \bar{h}_\nu(\omega) = h_\nu(\omega) - \frac{1}{E_{\nu 0}}. \tag{11}$$

For this manipulation, we make use of the commutation relation between current density and dipole density operators, and LWA is used for the vector potential. The details are given in sections 2.4 and 2.5 of [16].

The nonlocal relationship between induced current density and source EM field should be noted. In the microscopic scale, the cause $\boldsymbol{A}$ and the result $\boldsymbol{I}$ can occur at different positions, as far as they are within the extension of relevant wavefunctions. Since the coherence of matter wavefunction is important in nanostructures, this kind of susceptibility plays an essential role in the study of nanostructures.

The microscopic response is obtained by solving (10) with the microscopic M-eq

$$\frac{1}{c^2}\frac{\partial^2\boldsymbol{A}}{\partial t^2} - \nabla^2\boldsymbol{A} = \frac{4\pi}{c}\boldsymbol{I}_{\rm T}, \tag{12}$$

where $\boldsymbol{I}_{\rm T}$ is the transverse component of the induced current density. Though (10) contains the longitudinal (L) part, only the T component is necessary to obtain the self-consistent solution. Once it is obtained, one can use it in (10) to calculate the L component of $\boldsymbol{I}$.

Equation (12) is generally an integro-differential equation for $\boldsymbol{A}$, but it can be rewritten as a set of simultaneous linear equations of $\{F_{\nu 0}, F_{0\nu}\}$, because of the separable form of the microscopic susceptibility as an integral kernel. This rewriting facilitates the problem very much for practical applications. Since the microscopic structure of induced current density is included via the matrix elements of $\hat{\boldsymbol{I}}(\boldsymbol{r})$, this theory is quite suitable for the study of nanostructures. The details of this theory are given in [16] together with various examples of applications including nonlinear response. In this paper, we use this theory as a reliable basis for the derivation of macroscopic M-eqs as a new example of an application.

2.2. LWA of microscopic response theory

If the LWA is valid, the spatial variation of vector potential $\boldsymbol{A}(\boldsymbol{r},\omega)$ and induced current density $\boldsymbol{I}(\boldsymbol{r},\omega)$ will be weak in comparison with that of the matrix elements of the current density. Thus, the variables $\boldsymbol{A}$ and $\boldsymbol{I}$ are represented by their long wavelength components alone. The form of M-eqs for $\boldsymbol{A}$, (12), is kept unaltered under the LWA. In the Fourier representation, we have

$$\left(-\frac{\omega^2}{c^2} + k^2\right)\tilde{\boldsymbol{A}}(\boldsymbol{k},\omega) = \frac{4\pi}{c}\tilde{\boldsymbol{I}}_{\rm T}(\boldsymbol{k},\omega) \tag{13}$$

with the understanding that only small k components have appreciable amplitudes. A similar expression holds for the constitutive equation (10) as

$$\tilde{\boldsymbol{I}}(\boldsymbol{k},\omega) = \frac{1}{c}\sum_\nu [\bar{g}_\nu(\omega)\hat{\boldsymbol{I}}_{0\nu}(\boldsymbol{k})F_{\nu 0}(\omega) + \bar{h}_\nu(\omega)\hat{\boldsymbol{I}}_{\nu 0}(\boldsymbol{k})F_{0\nu}(\omega)]. \tag{14}$$

Here also, only small k components are considered to have appreciable amplitudes.

The factor $F_{\mu\nu}(\omega)$ can be rewritten as

$$F_{\mu\nu}(\omega) = \sum_{\boldsymbol{k}'}\tilde{\boldsymbol{I}}_{\mu\nu}(-\boldsymbol{k}')\cdot\tilde{\boldsymbol{A}}(\boldsymbol{k}',\omega), \tag{15}$$

which in general contains all the $\boldsymbol{k}'$-components. If LWA is valid, however, only small $\boldsymbol{k}'$ s make the central contribution, which is extracted as follows. For small $\boldsymbol{k}$, we may take the first few terms of the Taylor expansion (around $\bar{\boldsymbol{r}}$) as

$$\tilde{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{k}) = \frac{1}{V_{\rm n}}\int {\rm d}\boldsymbol{r}\,\exp[-{\rm i}\boldsymbol{k}\cdot\boldsymbol{r}]\hat{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{r}) = \frac{\exp(-{\rm i}\boldsymbol{k}\cdot\bar{\boldsymbol{r}})}{V_{\rm n}}(\bar{\boldsymbol{I}}_{\mu\nu} - {\rm i}\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{\mu\nu}) \tag{16}$$

where $\mu = 0$ or $\nu = 0$, $V_{\rm n}$ is the normalization volume, and

$$\bar{\boldsymbol{I}}_{\mu\nu} = \int {\rm d}\boldsymbol{r}\,\hat{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{r}), \qquad \bar{\boldsymbol{Q}}_{\mu\nu} = \int {\rm d}\boldsymbol{r}\,(\boldsymbol{r}-\bar{\boldsymbol{r}})\hat{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{r}) \tag{17}$$

represent the moments of the electric dipole (E1) and magnetic dipole (M1) (plus electric quadrupole (E2)) transitions, respectively. We choose $\bar{\boldsymbol{r}}$ for each transition '$\mu \leftrightarrow \nu$' in such a way (e.g. at the center of an impurity atom) that the moment $\bar{\boldsymbol{Q}}$ represents its physical meaning correctly. Then, the relation between $\tilde{\boldsymbol{I}}(\boldsymbol{k},\omega)$ and $\tilde{\boldsymbol{A}}(\boldsymbol{k}',\omega)$ obtained from (14) is generally not diagonal with respect to $\boldsymbol{k}$. However, if the macroscopic medium obtained by the LWA has translational symmetry, as usually anticipated, we may take the diagonal part $\tilde{\boldsymbol{I}}(\boldsymbol{k},\omega) = \chi_{\rm em}(\boldsymbol{k},\omega)\cdot\tilde{\boldsymbol{A}}(\boldsymbol{k},\omega)$, where

$$\chi_{\rm em}(\boldsymbol{k},\omega) = \sum_{\nu}\frac{N_\nu}{c}\big[\bar{g}_\nu(\omega)(\bar{\boldsymbol{I}}_{0\nu} - {\rm i}\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{0\nu})(\bar{\boldsymbol{I}}_{\nu0} + {\rm i}\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{\nu0}) + \bar{h}_\nu(\omega)(\bar{\boldsymbol{I}}_{\nu0} - {\rm i}\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{\nu0})(\bar{\boldsymbol{I}}_{0\nu} + {\rm i}\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{0\nu})\big]. \tag{18}$$

Here, we have replaced the factor $1/V_{\rm n}$ with the number density N_ν of localized states (of impurities, defects, etc) corresponding to the transition $0 \leftrightarrow \nu$, and the summation over ν is to be taken only once for the same localized transitions at different sites.

This is the general susceptibility of the present macroscopic scheme, and is the only susceptibility required to determine the complete (linear) response. It contains the contributions from both electric and magnetic dipole transitions, together with their mixing ($\boldsymbol{k}$-linear) terms. It should be noted that the mixing terms remain nonvanishing in the case of chiral symmetry, where each excited state $|\nu\rangle$ is active to both E1 and M1 transitions. In this case, we cannot properly define $\chi_{\rm e}$ and $\chi_{\rm m}$, or ϵ and μ. In the absence of chiral symmetry, on the other hand, E1 and M1 characters are not mixed, so that the k-linear terms vanish, and the susceptibility turns out to be a sum of E1 and M1 types of terms. This is the situation where we can use ϵ and μ, and only in this case, the dispersion equation of the present formulation coincides with the conventional one in terms of ϵ and μ, as will be discussed below in more detail.

2.3. Dispersion equation of plane waves

The macroscopic constitutive equation obtained above by LWA is described by the susceptibility tensor $\chi_{\rm em}(\boldsymbol{k},\omega)$. Substituting the expression of $\tilde{\boldsymbol{I}}(\boldsymbol{k},\omega)$ in the source term $(4\pi/c)\tilde{\boldsymbol{I}}$ of (13), we get the equation

$$\left(\frac{c^2k^2}{\omega^2} - 1\right)\tilde{A}_\xi = \frac{4\pi c}{\omega^2}\sum_\eta(\chi_{\rm em})_{\xi\eta}\tilde{A}_\eta \tag{19}$$

where ξ, η are the two Cartesian coordinate axes perpendicular to $\boldsymbol{k}$. The condition for the finite amplitude solution is the vanishing of the determinant of the coefficient (2×2) matrix, i.e.

$$\det\left|\frac{c^2k^2}{\omega^2} - 1 - \frac{4\pi c}{\omega^2}\chi_{\rm em}(\boldsymbol{k},\omega)\right| = 0. \tag{20}$$

This is the dispersion relation in the present scheme of macroscopic M-eqs. It should be compared with the conventional form of dispersion equation

$$\det\left|\frac{c^2k^2}{\omega^2} - \epsilon\mu\right| = 0, \tag{21}$$

where the T components of the tensors ϵ and μ should be inserted.

Apparently, the two dispersion equations are different, because the contributions of electric and magnetic polarizations appear as a product in (21), while in (20) as a sum (including an interference term). Moreover, the new result claims only one susceptibility, while there are two of them in the conventional formula. In view of the possible mixing of E1 and M1 transitions in the case of chiral symmetry, ϵ and μ can have common poles, which leads to an unphysical situation, i.e. the occurrence of second-order poles in the product $\epsilon\mu$ in spite of the linear response.

In the absence of chiral symmetry, however, E1 and M1 (+E2) transitions are grouped into different excited states. Namely, there is no excited state $|\nu\rangle$, making both $\bar{\boldsymbol{I}}_{\nu0}$ and $\bar{\boldsymbol{Q}}_{\nu0}$ nonzero. In this case, we may divide the ν summation into two groups as $(c/\omega)^2\chi_{\rm em} = \bar{\chi}_{\rm e} + \bar{\chi}_{\rm m}$, where $\bar{\chi}_{\rm e}$ and $\bar{\chi}_{\rm m}$ are the partial summations over ν for E1 and M1 (+E2) transitions, respectively, defined as

$$\bar{\chi}_{\rm e} = \frac{1}{\omega^2}\sum_\nu N_\nu[\bar{g}_\nu(\omega)\bar{\boldsymbol{I}}_{0\nu}\bar{\boldsymbol{I}}_{\nu0} + \bar{h}_\nu(\omega)\bar{\boldsymbol{I}}_{\nu0}\bar{\boldsymbol{I}}_{0\nu}], \tag{22}$$

$$\bar{\chi}_{\rm m} = \frac{k^2}{\omega^2}\sum_\nu N_\nu[\bar{g}_\nu(\omega)(\hat{\boldsymbol{k}}\cdot\bar{\boldsymbol{Q}}_{0\nu})(\hat{\boldsymbol{k}}\cdot\bar{\boldsymbol{Q}}_{\nu0}) + \bar{h}_\nu(\omega)(\hat{\boldsymbol{k}}\cdot\bar{\boldsymbol{Q}}_{\nu0})(\hat{\boldsymbol{k}}\cdot\bar{\boldsymbol{Q}}_{0\nu})] \tag{23}$$

for a unit vector $\hat{\boldsymbol{k}} = \boldsymbol{k}/|\boldsymbol{k}|$. In this case, the dispersion equation takes the form $\det|(c^2k^2/\omega^2) - (1+4\pi\bar{\chi}_{\rm e}+4\pi\bar{\chi}_{\rm m})| = 0$, which should be compared with the conventional form, (21), $\det|(c^2k^2/\omega^2) - (1+4\pi\chi_{\rm e})(1+4\pi\chi_{\rm m})| = 0$. It appears that, even in this simplified case, the two forms of dispersion equation are still different. However, if we use, instead of $\chi_{\rm m}$, the more fundamental magnetic susceptibility $\chi_{\rm B}$ defined by $\boldsymbol{M} = \chi_{\rm B}\boldsymbol{B}$ (see section 3.1 for more details), we have $\mu = 1+4\pi\chi_{\rm m} = (1-4\pi\chi_{\rm B})^{-1}$. Then, the conventional form can be rewritten as $\det|(c^2k^2/\omega^2) - (1+4\pi\chi_{\rm e}+4\pi\chi_{\rm B}c^2k^2/\omega^2)| = 0$, where the contributions of E1 and M1 transitions appear as a sum and the $\chi_{\rm B}$ term is multiplied with the factor of $\mathrm{O}(k^2)$. The relationship between $(\bar{\chi}_{\rm e}, \bar{\chi}_{\rm m})$ and $(\chi_{\rm e}, \chi_{\rm m})$ is obtained from the comparison of $\mathrm{O}(k^0)$ and $\mathrm{O}(k^2)$ terms of the two dispersion equations as

$$\chi_{\rm e} = \bar{\chi}_{\rm e}, \qquad \chi_{\rm B} = (\omega/ck)^2\bar{\chi}_{\rm m}. \tag{24}$$

In this way the equivalence of the new and conventional dispersion equations is demonstrated in the absence of chiral symmetry.

The above argument gives the answer to all the problems (i)–(iv) in the introduction. (i) Since we do not separate $\boldsymbol{I}$ into different components, there arises no problem of non-uniqueness. (ii) The electric and magnetic susceptibilities (χ_e and χ_B) correspond to the first-and second-order terms, respectively, of the LWA expansion of the microscopic susceptibility $\chi(\boldsymbol{r}, \boldsymbol{r}', \omega)$, in the absence of chiral symmetry. Namely, they are two tensors derived from a single nonlocal susceptibility. (iii) The (apparently) different wavenumber dependence of the two M1 transitions arises from the different ways of description, i.e. χ_m (or χ_B) in the case of spin resonance [6] and $\bar{\chi}_m$ in the case of orbital M1 transition [7]. (iv) The apparent difference in dispersion equation is reconciled by the use of χ_B instead of χ_m in the conventional description.

3. Discussions

3.1. Microscopic derivation of magnetic susceptibility

In order to derive χ_e and χ_m from the quantum mechanical calculation of $\boldsymbol{P}$ and $\boldsymbol{M}$ as linear response, we need an interaction term proportional, not to $\boldsymbol{A}$ as in (3), but to $\boldsymbol{E}$ and $\boldsymbol{H}$. An expression close to this can be properly obtained by making use of the fact that the addition of a total time derivative of arbitrary function (of time and position) does not affect the least action principle of the Lagrangian, i.e. we add the term $F(t) = (\mathrm{d}/\mathrm{d}t)\int \mathrm{d}\boldsymbol{r}\, \boldsymbol{P}\cdot\boldsymbol{A}/c$ to the Lagrangian (1) [2, 17]. The combination of this term with H_{int}, (3), leads to a new form of the interaction Hamiltonian as

$$H'_{\mathrm{int}} = -\int \mathrm{d}\boldsymbol{r}\,\{\boldsymbol{P}\cdot\boldsymbol{E}_{\mathrm{T}} + \boldsymbol{M}\cdot\boldsymbol{B}\}, \tag{25}$$

by the use of partial integration and $\boldsymbol{E}_{\mathrm{T}} = -(1/c)(\partial\boldsymbol{A}/\partial t)$, $\boldsymbol{B} = \nabla\times\boldsymbol{A}$.

The operator form of $\boldsymbol{P}$ and $\boldsymbol{M}$, satisfying the requirements $\nabla\cdot\boldsymbol{P} = -\rho$, $\boldsymbol{J} = (\partial\boldsymbol{P}/\partial t) + (1/c)\nabla\times\boldsymbol{M}$ (for charge neutral system) is known (section IV.C of [2]) as

$$\boldsymbol{P}(\boldsymbol{r}) = \int_0^1 \mathrm{d}u \sum_\ell e_\ell \boldsymbol{r}_\ell \delta(\boldsymbol{r} - u\boldsymbol{r}_\ell), \tag{26}$$

$$\boldsymbol{M}(\boldsymbol{r}) = \int_0^1 u\,\mathrm{d}u \sum_\ell e_\ell \boldsymbol{r}_\ell \times \boldsymbol{v}_\ell \delta(\boldsymbol{r} - u\boldsymbol{r}_\ell), \tag{27}$$

which enables us to calculate induced electric and magnetic polarizations with H'_{int} as a perturbation. The result of such a calculation should, however, be given as a functional of, not $\boldsymbol{E}$ and $\boldsymbol{H}$, but $\boldsymbol{E}$ and $\boldsymbol{B}$. Moreover, for the general case of symmetry, both $\boldsymbol{P}\cdot\boldsymbol{E}$ and $\boldsymbol{M}\cdot\boldsymbol{B}$ terms in H'_{int} should contribute to both of the induced polarizations, i.e. we should obtain $\boldsymbol{P}(\boldsymbol{E}, \boldsymbol{B})$ and $\boldsymbol{M}(\boldsymbol{E}, \boldsymbol{B})$. This does not lead to the usual definition of susceptibilities.

Only in the absence of chiral symmetry, where there is no mixing of E1 and M1 transitions, is $\boldsymbol{P}$ induced by $\boldsymbol{E}$ alone, and $\boldsymbol{M}$ by $\boldsymbol{B}$ alone, and this allows the definition of electric and magnetic susceptibility. In this case, however, a perturbation calculation similar to the one used for $\chi_{\mathrm{em}}(\boldsymbol{k}, \omega)$ (18), should give induced magnetization, not as $\boldsymbol{M} = \chi_m\boldsymbol{H}$, but as $\boldsymbol{M} = \chi_B\boldsymbol{B}$, since the interaction term (25) is linear in $\boldsymbol{B}$. The poles of χ_B correspond to the energies of magnetic excitations. Then, the definition $\boldsymbol{B} = \boldsymbol{H} + 4\pi\boldsymbol{M}$ and $\boldsymbol{M} = \chi_m\boldsymbol{H} = \chi_B\boldsymbol{B}$ leads to $\chi_m = \chi_B(1 - 4\pi\chi_B)^{-1}$ and $\mu = (1 - 4\pi\chi_B)^{-1}$. The last relation has provided an essential key to prove the equivalence of the two dispersion equations in section 2.3.

The use of χ_B as the linear response coefficient for magnetic excitations is a logical consequence from the first-principles Lagrangian, but there is so much literature using χ_m instead of χ_B. However, this paper is not the first one to claim the use of χ_B; see [15] for example. A proposal is made in section 3.5 to show an observable difference between the use of χ_m and χ_B for analysis.

3.2. Comparison with other schemes of macroscopic M-eqs

There have been several proposals of single susceptibility schemes of macroscopic M-eqs [8, 9, 14], which are apparently motivated by the argument of Landau–Lifshitz [4] that, as frequency increases, magnetization tends to lose its physical meaning, i.e. it becomes meaningless to separate current density into the contributions of $\boldsymbol{P}$ and $\boldsymbol{M}$. This is rather similar to our motivation, but ours is one step stronger. Namely, 'for any frequency region, we need only one vector field $\boldsymbol{I}$ as in microscopic M-eqs, and it should be possible to write macroscopic M-eqs and constitutive equations without separating $\boldsymbol{I}$ into the contributions of $\boldsymbol{P}$ and $\boldsymbol{M}$'.

In both of the proposals, they derive the microscopic forms of induced current density, from which one could derive conductivity and translate it into dielectric function. The calculation of current density is equivalent to that of this paper. As to the translation of conductivity into dielectric function, it is a standard matter in the absence of chiral symmetry, but it is questionable if one can do it also for the chiral case. The authors of [8] call their scheme the '$\boldsymbol{E}, \boldsymbol{D}, \boldsymbol{B}$' approach. In the same manner, we might call ours the '(macroscopic) $\boldsymbol{E}, \boldsymbol{B}$' approach. In [9], they describe the case of dipole approximation, which corresponds to our $\mathrm{O}(k^0)$ term of χ_{em}. Our results in section 2 show the importance of the whole set of '$\mathrm{O}(k^0), \mathrm{O}(k^1), \mathrm{O}(k^2)$' terms of the Taylor expansion to obtain the consistent picture of a single susceptibility scheme of macroscopic M-eqs free from the incompleteness problems (i)–(iv) in section 1. Both of [9] and [14] stress the importance of distinguishing the presence and absence of spatial dispersion within the macroscopic M-eqs, but we regard it simply as the different order terms of Taylor expansion within LWA. The '$\mathrm{O}(k^1), \mathrm{O}(k^2)$' terms do not increase the number of coupled (polariton) modes, so that this is not a type of spatial dispersion effect requiring additional boundary conditions. In spite of the similarity in motivation and parts of the formulas, neither of them show any systematic expansion of the microscopic constitutive equation to obtain a generalized form of macroscopic M-eqs. In this sense, we may claim that the present method and result represent a more complete level of single susceptibility scheme.

There is a different approach to macroscopic M-eqs by Nelson [18], where he applies LWA to the Lagrangian of a matter–EM field system, rewriting it into a 'continuum'

Lagrangian. The explicit use of LWA in the mathematical treatment is a common feature to the present theory, but the physical meaning is quite different. By the application of LWA to the Lagrangian, the dynamics of matter is described only by the long wavelength (LW) components, i.e. the LW eigenmodes of matter such as acoustic and optical phonons and excitons. Thus the only contribution to susceptibility is made from the LW modes of matter, i.e. the susceptibility has poles only at the frequencies of these LW modes. Since all the dynamical variables of short wavelength components are eliminated by the LWA of the Lagrangian, there is no chance for localized eigenstates of the matter to contribute to susceptibility. When we consider a problem, for example, of changing the refractive index of a material by adding impurities, the main change is caused by the localized excitations at the impurities. But they cannot be taken into account in Nelson's treatment, because they are not LW modes of matter. In contrast, our approach based on the LWA of microscopic constitutive equations takes all the contributions of the eigenmodes of matter according to their weights in LWA, i.e. oscillator strengths. Thus, our approach provides a continuous relationship between microscopic and macroscopic descriptions of the EM response, including the method to evaluate the validity condition of the LWA (see section 3.6).

3.3. Theoretical schemes for chiral symmetry

Having shown the equivalence of the new and conventional macroscopic M-eqs in the absence of chiral symmetry, we now make the comparison in the case of chiral symmetry between the new scheme and the existing one, the Drude–Born–Fedorov (DBF) constitutive equations, which extends the definition of ϵ and μ in such a way to allow the possibility of 'magnetic field induced $\boldsymbol{P}$' and 'electric field induced $\boldsymbol{M}$'. The form of DBF equations for a homogeneous isotropic case is [19]

$$\boldsymbol{D} = \epsilon(\boldsymbol{E} + \beta\nabla \times \boldsymbol{E}), \qquad \boldsymbol{B} = \mu(\boldsymbol{H} + \beta\nabla \times \boldsymbol{H}), \quad (28)$$

where the new parameter β (chiral admittance) takes care of chiral symmetry.

This set of equations allows us to describe, for example, different phase velocities of right and left circularly polarized lights for finite value of β. By solving the M-eqs $c\nabla \times \boldsymbol{H} = \partial\boldsymbol{D}/\partial t, c\nabla \times \boldsymbol{E} = -\partial\boldsymbol{B}/\partial t$ and the DBF equation simultaneously, we get the dispersion equation

$$\left(\frac{ck}{\omega}\right)^2 = \epsilon\mu\left(1 \pm \frac{\beta\omega}{c}\sqrt{\epsilon\mu}\right)^{-2}. \quad (29)$$

The $\pm$ sign in front of β describes the two possible values of k for a given ω, leading to different phase velocities.

This dispersion equation is obviously different from (20) with respect to their pole structure on the rhs. In fact, while the corresponding part of (20) is a superposition of single poles of $\chi_{\rm em}$ representing the excitation energies of matter, the rhs of (29) has a more complicated pole structure, i.e. higher-order poles and the poles not corresponding to matter excitation energies. Since E1 and M1 characters of matter excitations are mixed in chiral materials, the successful trick in section 2.3 to divide $\chi_{\rm em}$ into $\chi_{\rm e}$ and $\chi_{\rm B}$ in the absence of chiral symmetry fails in this case. Thus the DBF equations cannot be justified from a microscopic basis, i.e. we cannot obtain the quantum mechanical expressions of ϵ, μ, β consistent with $\chi_{\rm em}$. It is preferable to use $\chi_{\rm em}$ with the k-linear term to take care of the chiral nature, rather than the DBF equations.

Chiral symmetry of matter leads to the interference of electric and magnetic polarizations. It has been studied in various cases, such as the rotation of polarization plane [19], the mixing effect between electric and magnetic dipole characters, e.g. of the excitons in CdS [20] and the coupled 'Landau level–spin flip' transitions in GaAs [21], and the Jones effect in atomic spectroscopy [22]. Microscopic analysis of these effects, essentially corresponding to the microscopic nonlocal response in section 2, is shown in [20–22]. When we treat chiral symmetry in macroscopic response under resonant conditions, the argument of pole structure given above suggests we should avoid using the DBF equations.

3.4. Definition of left-handed materials (LHMs)

Considering that $\chi_{\rm e}$ and $\chi_{\rm B}$ can be properly defined only in the absence of chiral symmetry, we should reconsider the definition of LHMs by Veselago, i.e. $\epsilon < 0, \mu < 0$ [12], because LHM is possible also in chiral symmetry. For that purpose, it would be appropriate to use 'the occurrence of a dispersion branch with $v_{\rm ph} \times v_{\rm g} < 0$', where $v_{\rm ph}$ and $v_{\rm g}$ are phase and group velocities, respectively. This is one of the characteristics of LHM described by Veselago, but it has a more general applicability as a definition of LHM. LHM can be made of materials with or without chiral symmetry. Though it is recommended to use $\chi_{\rm em}$ in general, it is possible to use the conventional 'ϵ, μ' scheme in the absence of chiral symmetry under the condition of using $\chi_{\rm B}$ as the magnetic susceptibility.

The part of positive $v_{\rm g}$ in the negative k region is important because the plane wave on this part of the branch should be connected to the incident EM field via the boundary conditions at the interface. When two (or more) plane waves exist in the medium at the frequency of an incident wave, the general criterion for the correct choice of the branch is that the connected wave in the medium should not diverge as the size of matter becomes larger. From a general consideration of the dispersion curves in the complex (ω, k) plane [23], it is concluded that the choice of the branch with positive $v_{\rm g}$ meets this criterion. Physically, this means a simple fact that the plane wave with positive $v_{\rm g}$ decays in the direction of its propagation. Therefore, it applies to any dispersion curves of either right- or left-handed character.

3.5. Observable difference in using $\chi_{\rm m}$ or $\chi_{\rm B}$

We consider a LHM situation in the absence of chiral symmetry where a magnetic transition occurs in the frequency region where $1 + (4\pi c/\omega^2)\bar{\chi}_{\rm e}$ $(=\epsilon_{\rm b}) < 0$. Let us consider the two cases to assign the resonant frequency ω_0 of the magnetic transition: (A) $\chi_{\rm m} = b/(\omega_0 - \omega - {\rm i}0^+)$, (B) $\chi_{\rm B} = b/(\omega_0 - \omega - {\rm i}0^+)$, where b is the strength of the transition. The corresponding dispersion relations are (A) $(c^2k^2/\omega^2) = \epsilon_{\rm b}[1 + (4\pi b)/(\omega_0 - \omega - {\rm i}0^+)]$, (B) $(c^2k^2/\omega)^2 = \epsilon_{\rm b}/[1 -$

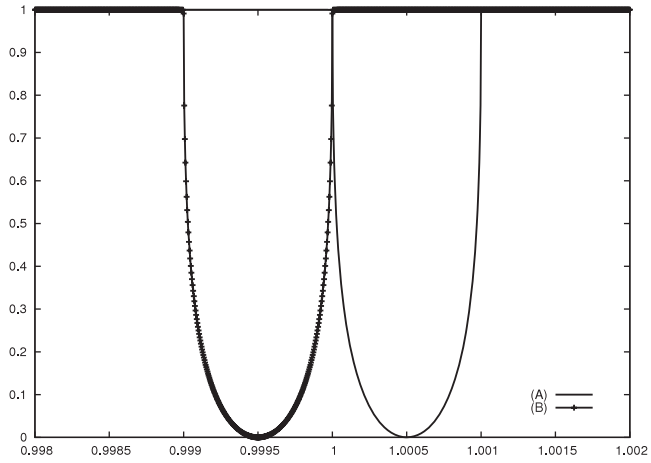

Figure 1. Reflectivity versus normalized frequency ω/ω_0, showing a transmission window in the total reflection range. The parameter values $\epsilon_b = -1$ and $4\pi b/\omega_0 = 0.001$ are used. Two curves for case (A) and (B) are almost identical, except for their positions with respect to the resonant frequency $\omega/\omega_0 = 1$.

$(4\pi b)/(\omega_0 - \omega - \mathrm{i}0^+)]$. The solution of this equation gives the dispersion relation $k = k(\omega)$ and refractive index $n = ck/\omega$, with $v_{\mathrm{ph}} \times v_{\mathrm{g}} < 0$. For the positive v_{g} branch of LHM behavior, k and n are negative. The reflectivity spectrum $R = |(n+1)/(n-1)|^2$ $(n < 0)$ of a semi-infinite medium for normal incidence is shown in figure 1. The remarkable point is the relative position of ω_0 with respect to the reflectivity dip due to the LHM branch. In (A), ω_0 occurs on the lower frequency end of the window, and in (B) on the higher frequency end. This could be checked experimentally, via an appropriate model system, e.g. a magnetic resonance of well-defined impurities overlapping with an E1 type phonon resonance.

3.6. Validity of LWA

In section 2.2, we have used the LWA, assuming its validity for systems under consideration. The validity condition of LWA is not provided by the macroscopic theory itself, but must be checked independently through the microscopic scheme in section 2, where all the wavelength components are included in the response field. Only when the amplitudes of the short wavelength components are small enough in comparison with the LW components, is the LWA a good approximation.

The validity condition of the LWA is closely connected with the resonant or non-resonant condition of the optical process in question. In resonant optical processes, only a few excited states will be resonant with incident frequency, and they will make the main contribution to the induced current density. Thus, the current density acquires a characteristic microscopic spatial structure reflecting the quantum mechanical wavefunctions of the resonant excited states. All the other non-resonant states make contributions of more or less comparable amplitudes, so that their superposition will have no particular microscopic structure. Therefore, non-resonant processes could generally be handled by a macroscopic scheme, and resonant processes should mostly be treated by microscopic theory. However, the resonant processes due to uniformly distributed well-localized impurity states could be treated by the macroscopic theory, as far as one considers the average properties of these localized states.

4. Summary

Applying LWA to the microscopic response of a matter–EM field system represented by a general Hamiltonian (including relativistic correction), we have derived new macroscopic M-eqs, which require only a single susceptibility $\chi_{\mathrm{em}}(\boldsymbol{k}, \omega)$ relating induced current density with vector potential. These M-eqs are more general than the '$\boldsymbol{E}$, $\boldsymbol{D}$, $\boldsymbol{B}$, $\boldsymbol{H}$' scheme. The conventional description by ϵ and μ is recovered in the absence of chiral symmetry, under the additional condition that one should use χ_{B} instead of χ_{m}. The phenomenology of DBF equations used for the description of chiral materials has been shown to be unjustifiable from this microscopic basis. The dispersion equation $\det|(ck/\omega)^2 - 1 - (4\pi c/\omega^2)\chi_{\mathrm{em}}| = 0$, is valid both in the presence and absence of chiral symmetry. The problems of uniqueness and consistency inherent to conventional M-eqs are solved by this new scheme. An observable difference in using χ_{B} or χ_{m} is demonstrated, and discussions about the comparison with existing theories are given.

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