Microscopic Expression of Chiral Susceptibilities

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Abstract

A first-principles derivation of susceptibilities including chiral ones is presented,and its relevance to the advanced study of metamaterials is discussed. Matter Hamiltonian and matter-EM field interaction are taken in a general form, which leads to microscopic, and then, via long wavelength approximation, macroscopic constitutive equations. The quantum mechanical expressions of susceptibilities thus obtained show restrictions to the modeling in metamaterials study.

1. Introduction

In an elementary argument of EM response of matter, electric and magnetic polarizations are induced by electric and magnetic fields, respectively. However, if the symmetry of matter is chiral, there are also electric (magnetic) polarization induced by magnetic (electric) field. In order to describe this situation as a macroscopic response, the usual relations $\boldsymbol{D} = \epsilon \boldsymbol{E},\ \boldsymbol{B} = \mu \boldsymbol{H}$ are extended to $\boldsymbol{D} = \epsilon[\boldsymbol{E} + \beta \nabla \times \boldsymbol{E}],\ \boldsymbol{B} = \mu[\boldsymbol{H} + \beta \nabla \times \boldsymbol{H}]$ [1] (case of isotropic uniform systems), which is called Drude-Born-Fedorov (DBF) constitutive equations [2]. This is a phenomenology based on symmetry arguments alone, and the chiral susceptibility β is regarded as a free parameter. There has been no theory to give a microscopic expression of this kind of parameter, to the knowledge of the author. Because of this situation, the extended constitutive equations appear in different, inequivalent forms in various literatures, which was the case also in Metamaterials 2010 at Karlsruhe. This seems to be tolerated as a phenomenology. From the logical point of view, however, they must have unique expressions, since all the EM response theories, from QED to semiclassical micro- and macroscopic theories, should belong to a hierarchy, where a lower rank theory is derived from a higher one via an approximation. This type of macroscopic response theory was recently made by the present author [3], which actually shows the microscopic expression of chiral susceptibilities.

Study of metamaterials seems to have been accelerated by a freedom in ascribing parameter values (and signs) to susceptibilities. In its mature level, however, it should be reminded that there exist certain restrictions to such a freedom due to the logically allowed relationship between micro- and macroscopic susceptibilities. In view of the new trend in metamaterials study, such as microscopic metamaterials, nonlocality, chirality, etc., it is timely to discuss this logically allowed relationship, as well as the validity of macroscopic description.

Since the new formulation of macroscopic response theory is not yet well-known, we give its outline first, and then, discuss its relevance to metamaterials study.

2. General forms of micro- and macroscopic susceptibilities

Starting from a general Lagrangian of interacting charge - EM field system in its standard form, we derive matter Hamiltonian and interaction Hamiltonian in unambiguous forms, which allows us to calculate

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induced current density due to source EM field. Within the framework of non-relativistic theory, it is possible to consider relativistic corrections, such as spin-orbit interaction, spin Zeeman interaction, etc. [4, 5]. Including the spin Zeeman interaction in interaction Hamiltonian $H_{\rm int}$ and the rest in matter Hamiltonian $H_{\rm M}$, we can express them as $H_{\rm M} = \sum_\ell (\boldsymbol{p}_\ell^2/)2m_\ell + U_{\rm C} + H_{\rm rel}$ and

$$H_{\rm int} = -\frac{1}{c}\int {\rm d}\boldsymbol{r}[\boldsymbol{J}_0(\boldsymbol{r}) + \boldsymbol{J}_{\rm spin}(\boldsymbol{r})]\cdot\boldsymbol{A}(\boldsymbol{r}) + \int {\rm d}\boldsymbol{r}\rho(\boldsymbol{r})\phi_{\rm ext}(\boldsymbol{r}) \tag{1}$$

in Coulomb gauge, where $U_{\rm C}$ is the Coulomb potential among charged particles, $H_{\rm rel}$ the relativistic corrections except spin Zeemen term, ρ the charge density, of the matter in consideration, $\phi_{\rm ext}$ the potential due to external charge density, $\boldsymbol{J}_0(\boldsymbol{r}) = \sum_\ell (e_\ell/2m_\ell)[\boldsymbol{p}_\ell\delta(\boldsymbol{r}-\boldsymbol{r}_\ell) + \delta(\boldsymbol{r}-\boldsymbol{r}_\ell)\boldsymbol{p}_\ell]$, and $\boldsymbol{J}_{\rm spin} = c\nabla\times\boldsymbol{M}^{\rm (spin)}$ for spin magnetization $\boldsymbol{M}^{\rm (spin)}$.

For a matter system initially in the ground state, we can calculate the microscopic current density linear in $\boldsymbol{A}$ and $\phi_{\rm ext}$ induced by $H_{\rm int}$ as

$$\boldsymbol{I}(\boldsymbol{r},\omega) = \int {\rm d}\boldsymbol{r}'\chi_{\rm cd}(\boldsymbol{r},\boldsymbol{r}';\omega)\cdot[\boldsymbol{A}(\boldsymbol{r}',\omega) + (c/i\omega)\boldsymbol{E}_{\rm extL}(\boldsymbol{r}',\omega)]\ , \tag{2}$$

where $\boldsymbol{E}_{\rm extL} = -\nabla\phi_{\rm ext}$, and $\chi_{\rm cd}(\boldsymbol{r},\boldsymbol{r}',\omega) = (1/c)\sum_\nu[\bar{g}_\nu(\omega)\boldsymbol{I}_{0\nu}(\boldsymbol{r})\boldsymbol{I}_{\nu 0}(\boldsymbol{r}') + \bar{h}_\nu(\omega)\boldsymbol{I}_{\nu 0}(\boldsymbol{r})\boldsymbol{I}_{0\nu}(\boldsymbol{r}')]$, $\bar{g}_\nu(\omega) = 1/(E_{\nu 0} - \hbar\omega - i0^+) - 1/E_{\nu 0}$, $\bar{h}_\nu(\omega) = 1/(E_{\nu 0} + \hbar\omega + i0^+) - 1/E_{\nu 0}$, where $\boldsymbol{I} = \boldsymbol{J}_0 + \boldsymbol{J}_{\rm spin}$, $H_{\rm M}|\nu\rangle = E_\nu|\nu\rangle$, $E_{\nu 0} = E_\nu - E_0$, E_0 being the ground state energy. The single vector $\boldsymbol{A} + (c/i\omega)\boldsymbol{E}_{\rm extL}$ takes care of the transverse (electric and magnetic) and longitudinal parts of source EM field. Equation (2) describes all the linear (electric, magnetic and chiral) response of matter for a general source field with electric and magnetic, as well as transverse and longitudinal, character. The microscopic EM response is obtained from the solution of eq.(2) coupled with the microscopic Maxwell equations $-\nabla^2\boldsymbol{A} - (\omega^2/c^2)\boldsymbol{A} = (4\pi/c)\boldsymbol{I}^{\rm (T)}$ [6] .

When the spatial extensions of the quantum mechanical excited states are small in comparison with the wavelength of the relevant EM field, a macroscopic description will be allowed. The long wavelength approximation of the $(\boldsymbol{k},\omega)$ Fourier component of eq.(2) leads to $\boldsymbol{I}(\boldsymbol{k},\omega) = \chi_{\rm em}(\boldsymbol{k},\omega)\cdot[\boldsymbol{A}(\boldsymbol{k},\omega) + (c/i\omega)\boldsymbol{E}_{\rm extL}(\boldsymbol{k},\omega)]$, with the macroscopic susceptibility $\chi_{\rm em}(\boldsymbol{k},\omega) = (1/c)\sum_\nu[\bar{g}_\nu(\omega)\tilde{\boldsymbol{I}}_{0\nu}(\boldsymbol{k})\tilde{\boldsymbol{I}}_{\nu 0}(-\boldsymbol{k}) + \bar{h}_\nu(\omega)\tilde{\boldsymbol{I}}_{\nu 0}(\boldsymbol{k})\tilde{\boldsymbol{I}}_{0\nu}(-\boldsymbol{k})]$. The definition of the matrix elements is given as

$$\tilde{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{k}) = (\exp(-i\boldsymbol{k}\cdot\bar{\boldsymbol{r}})/V)[\bar{\boldsymbol{J}}_{\mu\nu} - i\boldsymbol{k}\cdot\bar{\mathbf{Q}}_{\mu\nu}^{\rm (e2)} + ic\boldsymbol{k}\times\bar{\boldsymbol{M}}_{\mu\nu} + O(k^2)] \tag{3}$$

$$\bar{\boldsymbol{J}}_{\mu\nu} = \int {\rm d}\boldsymbol{r}\ \langle\mu|\boldsymbol{J}_0|\nu\rangle,\ \ \bar{\boldsymbol{M}}_{\mu\nu} = \bar{\boldsymbol{M}}_{\mu\nu}^{\rm (spin)} + \bar{\boldsymbol{M}}_{\mu\nu}^{\rm (orb)}\ , \tag{4}$$

$$\boldsymbol{k}\cdot\bar{\boldsymbol{Q}}_{\mu\nu}^{\rm (e2)} = \sum_\ell \frac{e_\ell}{2m_\ell}\int {\rm d}\boldsymbol{r}\ \{<\mu|(\boldsymbol{r}_\ell - \bar{\boldsymbol{r}})\boldsymbol{k}\cdot\boldsymbol{p}_\ell\ \delta(\boldsymbol{r}_\ell - \boldsymbol{r}) + \delta(\boldsymbol{r}_\ell - \boldsymbol{r})\ (\boldsymbol{r}_\ell - \bar{\boldsymbol{r}})\boldsymbol{k}\cdot\boldsymbol{p}_\ell|\nu>\}\ , \tag{5}$$

$$\bar{\boldsymbol{M}}_{\mu\nu}^{\rm (orb)} = \sum_\ell (e_\ell/2m_\ell c)\int {\rm d}\boldsymbol{r}\ <\mu|\boldsymbol{L}_\ell(\bar{\boldsymbol{r}})\ \delta(\boldsymbol{r}_\ell - \boldsymbol{r}) + \delta(\boldsymbol{r}_\ell - \boldsymbol{r})\ \boldsymbol{L}_\ell(\bar{\boldsymbol{r}})|\nu>\ , \tag{6}$$

where $\boldsymbol{L}_\ell(\bar{\boldsymbol{r}}) = (\boldsymbol{r}_\ell - \bar{\boldsymbol{r}})\times\boldsymbol{p}_\ell$, V is the quantization volume of $\boldsymbol{k}$, and $\bar{\boldsymbol{r}}$ is the center coordinate to make Taylor expansion of $\tilde{\boldsymbol{I}}_{\mu\nu}(\boldsymbol{k})$. Noting that the matrix elements $\bar{\boldsymbol{J}}_{\mu\nu}, \bar{\mathbf{Q}}_{\mu\nu}^{\rm (e2)}, \bar{\boldsymbol{M}}_{\mu\nu}$ are nonzero for electric dipole, electric quadrupole and magnetic dipole transitions, respectively, we see how the symmetry of a given matter system affects the form of $\chi_{\rm em}$. When they belong to different irreducible representaions of the group in consideration, there is no $\boldsymbol{k}$-linear term, i.e., the system is non-chiral. In a chiral system, we need a lowering of symmetry which causes the mixing of these matrix elements for same excited states, leading to the $\boldsymbol{k}$-linear terms in $\chi_{\rm em}$.

The decomposition of current density $\boldsymbol{I} = (\partial\boldsymbol{P}/\partial t) + c\nabla\times\boldsymbol{M}$ holds as both operators and expectation values [7]. Using this, we can rewrite the $(\boldsymbol{k},\omega)$ Fourier component of eq.(2) into the form

$\tilde{\boldsymbol{I}} = -i\omega(\tilde{\boldsymbol{P}}_{\rm E} + \tilde{\boldsymbol{P}}_{\rm B}) + ic\boldsymbol{k} \times (\tilde{\boldsymbol{M}}_{\rm E} + \tilde{\boldsymbol{M}}_{\rm B})$, where the generalized polarizations are defined as $\tilde{\boldsymbol{P}}_{\rm E} = \chi_{\rm eE}\tilde{\boldsymbol{E}}, \tilde{\boldsymbol{P}}_{\rm B} = \chi_{\rm eB}\tilde{\boldsymbol{B}}, \tilde{\boldsymbol{M}}_{\rm E} = \chi_{\rm mE}\tilde{\boldsymbol{E}}, \tilde{\boldsymbol{M}}_{\rm B} = \chi_{\rm mB}\tilde{\boldsymbol{B}}$ with the quantum mechanical expressions of the new susceptibilities

$$\begin{aligned} \chi_{\rm eE} &= \frac{1}{\omega^2 V}\sum_\nu [\bar{g}_\nu(\bar{\boldsymbol{J}}_{0\nu} - i\boldsymbol{k}\cdot\bar{\mathbf{Q}}^{(\rm e2)}_{0\nu})(\bar{\boldsymbol{J}}_{\nu 0} + i\boldsymbol{k}\cdot\bar{\mathbf{Q}}^{(\rm e2)}_{\nu 0}) \\ &\qquad + \bar{h}_\nu(\bar{\boldsymbol{J}}_{\nu 0} - i\boldsymbol{k}\cdot\bar{\mathbf{Q}}^{(\rm e2)}_{\nu 0})(\bar{\boldsymbol{J}}_{0\nu} + i\boldsymbol{k}\cdot\bar{\mathbf{Q}}^{(\rm e2)}_{0\nu})]\,, \qquad (7) \\ \chi_{\rm eB} &= \frac{i}{\omega V}\sum_\nu [\bar{g}_\nu(\bar{\boldsymbol{J}}_{0\nu} - i\boldsymbol{k}\cdot\bar{\mathbf{Q}}^{(\rm e2)}_{0\nu})\bar{\boldsymbol{M}}_{\nu 0} + \bar{h}_\nu(\bar{\boldsymbol{J}}_{\nu 0} - i\boldsymbol{k}\cdot\bar{\mathbf{Q}}^{(\rm e2)}_{\nu 0})\bar{\boldsymbol{M}}_{0\nu}]\,, \qquad (8) \\ \chi_{\rm mB} &= \frac{1}{V}\sum_\nu [\bar{g}_\nu\bar{\boldsymbol{M}}_{0\nu}\bar{\boldsymbol{M}}_{\nu 0} + \bar{h}_\nu\bar{\boldsymbol{M}}_{\nu 0}\bar{\boldsymbol{M}}_{0\nu}]\,, \qquad (9) \\ \chi_{\rm mE} &= \frac{-i}{\omega V}\sum_\nu [\bar{g}_\nu\bar{\boldsymbol{M}}_{0\nu}(\bar{\boldsymbol{J}}_{\nu 0} + i\boldsymbol{k}\cdot\bar{\mathbf{Q}}^{(\rm e2)}_{\nu 0}) + \bar{h}_\nu\bar{\boldsymbol{M}}_{\nu 0}(\bar{\boldsymbol{J}}_{0\nu} + i\boldsymbol{k}\cdot\bar{\mathbf{Q}}^{(\rm e2)}_{0\nu})]\,. \qquad (10) \end{aligned}$$

The susceptibilities $\chi_{\rm eB}$ and $\chi_{\rm mE}$ represent the chiral nature of the system. This rewriting provides four constitutive equations, but they are not independent from one another. They can always be put together to reproduce the single equation (2). In terms of $\boldsymbol{D} = \boldsymbol{E} + 4\pi\boldsymbol{P}, \boldsymbol{H} = \boldsymbol{B} - 4\pi\boldsymbol{M}$, the equations given above take the form $\boldsymbol{D} = (1 + 4\pi\chi_{\rm eE})\boldsymbol{E} + 4\pi\chi_{\rm eB}\boldsymbol{B}\ , \quad \boldsymbol{H} = (1 - 4\pi\chi_{\rm mB})\boldsymbol{B} - 4\pi\chi_{\rm mE}\boldsymbol{E}$, which are not equivalent to the DBF equations [1].

In nonchiral systems, $\chi_{\rm eB}$ and $\chi_{\rm mE}$ are zero, and there is no common pole between $\chi_{\rm eE}$ and $\chi_{\rm mB}$. It should also be noted that the magnetic susceptibility is defined as $\tilde{\boldsymbol{M}}_{\rm B} = \chi_{\rm mB}\tilde{\boldsymbol{B}}$. The poles of $\chi_{\rm mB}$ are the excitation energies of $H_{\rm M}$ as given above, while those of the usual magnetic susceptibility $\chi_{\rm m}$ are not, since $\mu = 1 + 4\pi\chi_{\rm m} = 1/(1 - 4\pi\chi_{\rm mB})$. In a chiral system, it is important to note that the four susceptibilities $\chi_{\rm eE}, \chi_{\rm eB}, \chi_{\rm mE}, \chi_{\rm mB}$ have common poles in general. Therefore, we need a special care in considering resonant effect in chiral symmetry. It would be too naive to ascribe resonant behavior only to the k-linear part of the susceptibility, which is usually considered to represent chiral effect. One would need a consistent model to describe such a case from a microscopic level.

LWA is not always valid. If some of the induced polarization has coherence length longer than the wavelength of relevant EM field, we need to solve the problem as a microscopic response. If the remaining part of polarization has resonance due to macroscopic geometry (cavity effect), this will lead to a mixed micro- and macroscopic response. To handle this kind of problem, we divide $\chi_{\rm cd}$ into the resonant part due to the long coherence mode and the remaining part. Defining the background EM field by renormalizing the effect of the latter, we could solve the microscopic response due to the former for this renormalized macroscopic EM field by using [6].

In conclusion, the microscopic expression of chiral susceptibilities is presented, and its meaning is discussed in view of the application to advanced metamaterials study.

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