General separability of linear and nonlinear optical susceptibilities

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It is shown that all components of the electric susceptibilities act as separable integral kernels in the nonlocal theory of radiation-matter interaction. This reduces the problem of solving the self-consistent motions of the radiation and matter to a set of polynomial equations for a given order of nonlinearity.

I. INTRODUCTION

In the usual semiclassical treatment of interaction of radiation with matter, one solves Maxwell's equations, which contain the polarization $\mathbf{P}(\mathbf{r},t)$ defined as a function of space time $(\mathbf{r},t)$. This term plays the role of source field in the coupled differential equations. Since the polarization is induced by the electric field to be solved, another equation called "constitutive equation," taking the form

$$\mathbf{P}(\mathbf{r},\omega) = \int d\mathbf{r}'\chi^{(1)}(\mathbf{r},\mathbf{r}';\omega)\mathbf{E}(\mathbf{r}',\omega) + \sum_{\omega_1+\omega_2=\omega}\int d\mathbf{r}_1\int d\mathbf{r}_2\chi^{(2)}(\mathbf{r},\mathbf{r}_1,\mathbf{r}_2;\omega_1,\omega_2) \times\mathbf{E}(\mathbf{r}_1,\omega_1)\mathbf{E}(\mathbf{r}_2,\omega_2)+\cdots, \tag{1.1}$$

is set up for the temporal Fourier transforms. Here, the induced polarization $\mathbf{P}$ is described as a sum of integrals of various orders in the products of the electric field $\mathbf{E}(\mathbf{r},\omega)$. The relationship between $\mathbf{P}$ and $\mathbf{E}$ is nonlocal in the sense that $\mathbf{P}(\mathbf{r},\omega)$ is induced by $\mathbf{E}$, not only at $\mathbf{r}$, but also at all the points $\mathbf{r}'$, where the integral kernels are nonzero. These kernels are called linear and nonlinear polarizabilities or susceptibilities, and can be calculated from the first principles, as shown later. Their position dependences are determined by the wave functions of the excited states of matter.

In usual optical response theory of solids, which deals with the macroscopic electromagnetic (EM) field, the susceptibilities are approximated as local quantities. Namely, $\mathbf{P}(\mathbf{r},\omega)$ is assumed to be induced only by the field at the same position $\mathbf{E}(\mathbf{r},\omega)$. This type of theory has been quite successful in various aspects of radiation-matter interaction in solids and is regarded as the standard theory of optical responses.[1] However, with the recent appearance of mesoscopic or nano-scale objects as reproducible samples of physical investigations, it has become essential to study the details of size quantized energy levels and the corresponding wave functions. This means that we should keep the nonlocal nature of the susceptibilities as required from the first principles calculation.

At first glance, it seems a formidable task to take full account of the nonlocal nature, even in linear response, in solving Maxwell equations, which is reduced to an integro-differential equation of $\mathbf{E}(\mathbf{r},t)$. However, when the integral kernels are separable with respect to the integral variables, the integro-differential equation turns out to be a set of polynomial equations, which is certainly easier to solve. The separability is obvious for the resonant part of susceptibility, which is contributed from the $\mathbf{p}\cdot\mathbf{A}$ type term of the interaction Hamiltonian. This type of approach has been taken by us in various problems of linear and nonlinear optical responses, and the consequences of nonlocality have been demonstrated in the form of the "size, shape, internal-structure" dependence of resonant response of mesoscopic systems.[2–4]

The purpose of this paper is to extend the range of validity of the separable nature of susceptibilities. It will be shown that any components of susceptibilities, either resonant or nonresonant, are separable kernels. This argument will complete the formulation of nonlocal response theory, which brings the equations of interacting radiation-matter system as the Nth order nonlinear response into a set of polynomial equations of Nth order in general.

II. SEPARABILITY OF OPTICAL SUSCEPTIBILITIES

In order to make the following argument as general as possible within the semiclassical framework, we will pursue the self-consistent motions of radiation field and matter as those of vector potential $\mathbf{A}$ and current density $\mathbf{j}$, instead of $\mathbf{E}$ and $\mathbf{P}$ mentioned above.

The fundamental equations of nonlocal response theory[2] consist of two functional equations for the ω-Fourier components of $\mathbf{A}$ and $\mathbf{j}$ as

$$\tilde{\mathbf{A}}(\mathbf{r},\omega) = \tilde{\mathbf{A}}_0(\mathbf{r},\omega) + \mathcal{G}[\tilde{\mathbf{j}}]\ , \tag{2.1}$$

$$\tilde{\mathbf{j}}(\mathbf{r},\omega) = \mathcal{F}[\tilde{\mathbf{A}}]\ . \tag{2.2}$$

The first equation is the solution of the (microscopic) Maxwell's equations with the given source terms including charge and current densities. We choose the Coulomb gauge, $\mathrm{div}\mathbf{A} = 0$, by which the scalar potential is determined by the instantaneous charge density. Due to the continuity equation, the charge density can be elimi-

nated, and the solution is written in terms of the current density alone. The free field $\tilde{\mathbf{A}}_0$ usually represents the incident light. The form of the functional $\mathcal{G}$ is rather simple, and is given as

$$\mathcal{G}[\tilde{\mathbf{j}}] = \frac{1}{c}\int d\mathbf{r}' G_q(\mathbf{r}-\mathbf{r}')\tilde{\mathbf{j}}(\mathbf{r}',\omega) \ , \tag{2.3}$$

where

$$G_q(\mathbf{r}-\mathbf{r}') = (1+q^{-2}\text{grad div})\frac{e^{iq|\mathbf{r}-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{r}'|} - g^{-2}\ \text{grad div}\ \frac{1}{|\mathbf{r}-\mathbf{r}'|} \ , \tag{2.4}$$

and $q=\omega/c$. It is on the second equation, (2.2), that we want to put the main stress in this paper. The functional $\mathcal{F}$ is generally written in a similar form as Eq. (1.1), and we will show that the integral kernels are all separable.

The current density induced by a given field $\mathbf{A}(\mathbf{r},t)$ is calculated from first principles in the following way. The nonrelativistic Hamiltonian of a system of charged particles in the given vector potential $\mathbf{A}(\mathbf{r},t)$ and scalar potential $\phi(r,t)$ can be written as a sum of the following one particle Hamiltonian:

$$H = \sum_\ell \left\{ \frac{1}{2m_\ell}\left(\mathbf{p}_\ell - \frac{e_\ell}{c}\mathbf{A}(\mathbf{r}_\ell,t)\right)^2 + e_\ell\phi(\mathbf{r}_\ell)\right\} \ , \tag{2.5}$$

where $(\mathbf{r}_\ell,\mathbf{p}_\ell)$ and (e_ℓ,m_ℓ) are the (coordinate, momentum) and the (charge, mass) of the ℓth particle, and c is the velocity of light in vacuum. If we choose the Coulomb gauge, $\text{div}\mathbf{A}=0$, the scalar potential represents the instantaneous Coulomb interaction among all the charged particles. The Hamiltonian of the material system, H_0, is the $\mathbf{A}$-independent part of Eq. (2.5):

$$H_0 = H|_{\mathbf{A}=0} \ . \tag{2.6}$$

The scalar potential is regarded as a part of the matter Hamiltonian, which fits well to the usual understanding of matter in nonrelativistic regime. We refer to the eigenenergies and the corresponding eigenstates of H_0 by $\{E_\lambda\}$ and $\{|\lambda\rangle\}$, which will be used to derive some relations between matrix elements.

The Hamiltonian of the interaction of the radiation with matter is defined as the $\mathbf{A}$-dependent terms in H, namely,

$$H_{\text{int}}(t) = \sum_\ell \left(-\frac{e_\ell}{m_\ell c}\mathbf{A}_\ell\cdot\mathbf{p}_\ell + \frac{e_\ell^2}{2m_\ell c^2}\mathbf{A}_\ell^2\right) \ , \tag{2.7}$$

where $\mathbf{A}_\ell = \mathbf{A}(\mathbf{r}_\ell,t)$. (Since we treat $\mathbf{A}$ as classical variables, H_{int} depends on t explicitly even in the Schrödinger picture for the material system.) For a given field $\mathbf{A}(\mathbf{r},t)$, the time evolution of the matter system is described by the Liouville equation of matter density matrix ρ,

$$i\hbar\frac{d}{dt}\rho = [H_0+H_{\text{int}},\rho] \ . \tag{2.8}$$

The expectation value of the current density at $(\mathbf{r},t)$ is given by

$$\mathbf{j}(\mathbf{r},t) = \text{Tr}\{\rho\hat{\mathbf{J}}\} \ , \tag{2.9}$$

where $\hat{\mathbf{J}}(\mathbf{r},t)$ is the current density operator defined by

$$\hat{\mathbf{J}}(\mathbf{r},t) = \sum_\ell \left\{ \frac{e_\ell}{2m_\ell}[\mathbf{p}_\ell\delta(\mathbf{r}-\mathbf{r}_\ell)+\delta(\mathbf{r}-\mathbf{r}_\ell)\mathbf{p}_\ell] - \frac{e_\ell^2}{m_\ell c}\mathbf{A}(\mathbf{r},t)\delta(\mathbf{r}-\mathbf{r}_\ell)\right\} \ . \tag{2.10}$$

Its t dependence arises from the motion of the particles and $\mathbf{A}(\mathbf{r},t)$. It should be noted that this definition satisfies the continuity equation,

$$\frac{\partial}{\partial t}\hat{N} + \text{div}\hat{\mathbf{J}} = 0 \ , \tag{2.11}$$

where the charge density operator $\hat{N}(\mathbf{r})$ is defined as

$$\hat{N}(\mathbf{r},t) = \sum_\ell e_\ell\delta(\mathbf{r}-\mathbf{r}_\ell) \ . \tag{2.12}$$

Equation (2.11) can be derived by the direct time differentiation of Eq. (2.12) and the use of the Heisenberg equation of motion.

For later use, we note another relationship between $\hat{\mathbf{J}}$ and the "dipole density" defined by

$$\hat{\mathbf{R}}(\mathbf{r},t) = \sum_\ell e_\ell\mathbf{r}\delta(\mathbf{r}-\mathbf{r}_\ell) \ . \tag{2.13}$$

The time derivative of this $\hat{\mathbf{R}}$ is related to $\hat{\mathbf{J}}$ by

$$\frac{d}{dt}\hat{\mathbf{R}} = \frac{1}{i\hbar}[\hat{\mathbf{R}},H] = \hat{\mathbf{J}} \ ; \tag{2.14}$$

also, the following commutation relation holds for the (ξ,η) Cartesian components.

$$[\hat{R}_\xi(\mathbf{r},t),\hat{J}_\eta(\mathbf{r}',t)] = i\hbar\delta_{\xi\eta}\delta(\mathbf{r}-\mathbf{r}')\bar{N}(\mathbf{r},t) \ , \tag{2.15}$$

where

$$\bar{N}(\mathbf{r},t) = \sum_\ell \frac{e_\ell^2}{m_\ell}\delta(\mathbf{r}-\mathbf{r}_\ell) \ . \tag{2.16}$$

If we denote the current density operator in the absence of $\mathbf{A}$ as $\hat{\mathbf{I}}$, similar relations hold for $\hat{\mathbf{R}}$ and $\hat{\mathbf{I}}$ as

$$[\hat{\mathbf{R}}(\mathbf{r},t),H_0] = i\hbar\hat{\mathbf{I}}(\mathbf{r},t), \tag{2.17}$$

$$[\hat{R}_\xi(\mathbf{r},t),\hat{I}_\eta(\mathbf{r}',t)] = i\hbar\delta_{\xi\eta}\delta(\mathbf{r}-\mathbf{r}')\bar{N}(\mathbf{r},t) \ . \tag{2.18}$$

The t dependences of the operators $\hat{\mathbf{R}}$, $\hat{\mathbf{I}}$, and $\bar{N}$ arise only from the particles motion, so that we omit the argument t in the following. These two equations will be used later for the proof of separability.

Now, the solution of the Liouville equation (2.8) is obtained by a perturbation expansion with respect to H_{int}; the result is formally written as[5]

$$\rho(t) = \exp(-iH_0t/\hbar)\rho^I(t)\exp(iH_0t/\hbar) \ , \tag{2.19}$$

$$\rho^I(t) = U(t)\rho_0U^\dagger(t) \ , \tag{2.20}$$

$$U(t) = T\exp\left[-\frac{i}{\hbar}\int_{-\infty}^{t}dt'H_{\text{int}}^I(t')\right] \ , \tag{2.21}$$

where ρ_0 is the initial density matrix at $t = -\infty$, T is the usual time ordering operator, and the interaction representation of the operator $H_{\rm int}^I$ is defined as

$$H_{\rm int}^I(t) = \exp[iH_0 t/\hbar] H_{\rm int}(t) \exp[-iH_0 t/\hbar] \ . \quad (2.22)$$

As to the $\mathbf{A}$ dependence of the current density (2.9), there are three different sources, namely, $\mathbf{A}\cdot\mathbf{p}$ and $\mathbf{A}^2$ terms in $H_{\rm int}$, and the $\mathbf{A}$ dependent term in the operator $\hat{\mathbf{J}}$, (2.10).

The expansion of the exponential factor in (2.22) leads to the expression of $\rho(t)$ as a sum of linear and nonlinear terms with respect to $\mathbf{A}$. Each term contains a product of integral number of $\mathbf{A}$'s as an integrand over multiple space-time variables. The use of this form of ρ in the evaluation of $\mathbf{j}(\mathbf{r},t)$, (2.9), leads to a similar expression of $\mathbf{j}$ as a power expansion form with respect to $\mathbf{A}$'s under integral signs. Since each factor of $\mathbf{A}$ (or $\mathbf{A}^2$) originates from one of the three sources mentioned above, we need to examine the matrix elements of the operators $\sum(e_\ell/m_\ell c)\mathbf{p}_\ell\cdot\mathbf{A}_\ell$, $\sum(e_\ell^2/m_\ell c^2)\mathbf{A}_\ell^2$, and $\sum(e_\ell^2/m_\ell c)\mathbf{A}_\ell$, with respect to the many particle eigenfunctions of H_0. For this purpose, it is useful to rewrite them as

$$\sum_\ell \frac{e_\ell}{m_\ell c}\mathbf{p}_\ell\cdot\mathbf{A}(\mathbf{r}_\ell,t) = \frac{1}{c}\int \hat{\mathbf{I}}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r},t)d\mathbf{r}, \quad (2.23)$$

$$\sum_\ell \frac{e_\ell^2}{m_\ell c^2}\mathbf{A}(\mathbf{r}_\ell,t)^2 = \frac{1}{c^2}\int \bar{N}(\mathbf{r})\mathbf{A}(\mathbf{r},t)^2 d\mathbf{r}, \quad (2.24)$$

$$\sum_\ell \frac{e_\ell^2}{m_\ell c}\mathbf{A}(\mathbf{r}_\ell,t) = \frac{1}{c}\int \bar{N}(\mathbf{r})\mathbf{A}(\mathbf{r},t)d\mathbf{r} \ . \quad (2.25)$$

Since $\mathbf{A}(\mathbf{r},t)$ is a c number in the semiclassical framework, we only need to consider the matrix elements of the two operators $\hat{\mathbf{I}}(\mathbf{r})$ and $\bar{N}(\mathbf{r})$.

Now we denote the matrix element of the ω-Fourier component of Eq. (2.23) as

$$F_{\mu\nu}(\omega) = \int d\mathbf{r}\langle\mu|\hat{\mathbf{I}}(\mathbf{r})|\nu\rangle\cdot\tilde{\mathbf{A}}(\mathbf{r},\omega) \ , \quad (2.26)$$

where $\tilde{\mathbf{A}}(\mathbf{r},\omega)$ is the ω-Fourier component of $\mathbf{A}(\mathbf{r},t)$. We take the eigenstates of H_0 as state vectors to form the matrix elements. Then, the matrix elements (2.24) and (2.25) can be written in terms of $\{F\}$, by the help of (2.17) and (2.18), as

$$\int d\mathbf{r}\langle\mu|\bar{N}(\mathbf{r})|\nu\rangle\tilde{\mathbf{A}}(\mathbf{r},\omega)\cdot\tilde{\mathbf{A}}(\mathbf{r},\omega')$$

$$= \sum_\tau\left(\frac{F_{\mu\tau}(\omega)F_{\tau\nu}(\omega')}{E_{\tau\mu}} + \frac{F_{\mu\tau}(\omega')F_{\tau\nu}(\omega)}{E_{\tau\nu}}\right), \quad (2.27)$$

$$\langle\mu|\bar{N}(\mathbf{r})|\nu\rangle\tilde{\mathbf{A}}(\mathbf{r},\omega) = \sum_\tau\left(\frac{F_{\mu\tau}\mathbf{I}_{\tau\nu}(\mathbf{r})}{E_{\tau\nu}} + \frac{\mathbf{I}_{\mu\tau}(\mathbf{r})F_{\tau\nu}}{E_{\tau\mu}}\right) \ , \quad (2.28)$$

where $\mathbf{I}_{\mu\nu}(\mathbf{r}) = \langle\mu|\hat{\mathbf{I}}(\mathbf{r})|\nu\rangle$ and $E_{\tau\mu} = E_\tau - E_\mu$. Altogether, all the matrix elements of the three operators (2.23)–(2.25) are found to be linear or quadratic forms of F's. The current density is calculated by further carrying out time integrals of various products of such matrix elements, which yield frequency-dependent coefficients. Since the $\mathbf{A}$ dependence of the current density occurs only through these three types of matrix elements, we can conclude that $\mathbf{j}(\mathbf{r},t)$, (2.9), is written as a polynomial series of F's. In other words, the susceptibility functions of all orders are separable integral kernels.

Substituting the expression of $\mathbf{j}(\mathbf{r},t)$ in Eq. (2.1), we can obtain $\mathbf{A}(\mathbf{r},t)$ as a polynomial series of F's. Inserting this result in Eq. (2.26), we get a self-consistent set of polynomial equations for F's with various combinations of (μ,ν) and frequencies. The solution of this set of equations provides all the necessary quantities of the interacting radiation-matter system in question. In practice, we often restrict ourselves to a given order of nonlinearity, for example, to the Nth order nonlinear processes. Then, the equations to be solved are Nth order polynomial equations, which could be solved numerically.

III. DISCUSSION

In the previous section, we have shown that the susceptibility functions for the current density $\mathbf{j}(\mathbf{r},t)$ are separable integral kernels at any order and the integro-differential equation is reduced to a set of polynomial equations. If we confine the argument to the case of linear response, we can give a very simple and general expression of the self-consistent solution. To first order in $\mathbf{A}$, $\mathbf{j}$ is obtained as

$$\tilde{\mathbf{j}}(\mathbf{r},\omega) = \frac{1}{c}\sum_{\lambda\sigma\tau}\left\{F_{\lambda\sigma}(\omega-\omega_{\sigma\tau})\left(\frac{1}{E_{\lambda\tau}-\hbar\omega-i\gamma} - \frac{1}{E_{\lambda\tau}}\right)\mathbf{I}_{\tau\lambda}(\mathbf{r})\langle\sigma|\rho_0|\tau\rangle\right.$$
$$\left. -F_{\sigma\lambda}(\omega-\omega_{\tau\sigma})\left(\frac{1}{E_{\tau\lambda}-\hbar\omega-i\gamma} - \frac{1}{E_{\tau\lambda}}\right)\mathbf{I}_{\lambda\tau}(\mathbf{r})\langle\tau|\rho_0|\sigma\rangle\right\} \ , \quad (3.1)$$

where $\hbar\omega_{\lambda\tau} = E_{\lambda\tau}$ and γ is an infinitesimal parameter for adiabatic switching. The terms without ω in the energy denominator come from the term proportional to $\mathbf{A}$ in $\hat{\mathbf{J}}$. The procedure mentioned above gives the self-consistent equations for the F's as follows:

$$F_{\mu\nu}(\omega) = F_{\mu\nu}^{(0)}(\omega) + \sum_{\lambda\sigma\tau}[A_{\mu\nu,\tau\lambda}\langle\sigma|\rho_0|\tau\rangle F_{\lambda\sigma}(\omega-\omega_{\sigma\tau})$$
$$-A_{\mu\nu,\lambda\tau}\langle\tau|\rho_0|\sigma\rangle F_{\sigma\lambda}(\omega-\omega_{\tau\sigma})] \ , \quad (3.2)$$

where $F_{\mu\nu}^{(0)}(\omega)$ is defined in terms of the incident electro-

magnetic field by

$$F^{(0)}_{\mu\nu}(\omega) = \int d\mathbf{r}\mathbf{I}_{\mu\nu}(\mathbf{r}) \cdot \tilde{\mathbf{A}}_0(\mathbf{r},\omega) \ . \tag{3.3}$$

The coefficients A's represent the retarded interaction among the induced current density, and are given by

$$A_{\mu\nu,\tau\lambda} = \frac{1}{c^2}\left(\frac{1}{E_{\lambda\tau} - \hbar\omega} - \frac{1}{E_{\lambda\tau}}\right) \times \int d\mathbf{r} \int d\mathbf{r}' \mathbf{I}_{\mu\nu}(\mathbf{r}) G_q(\mathbf{r}-\mathbf{r}') \mathbf{I}_{\tau\lambda}(\mathbf{r}') \ , \tag{3.4}$$

where the dyadic Green's function G_q has been given by Eq. (2.4).

In particular, if the system initially stays at the ground state g, the above expressions become further simpler. Using $\rho_0 = |g\rangle\langle g|$, we obtain

$$\tilde{\mathbf{j}}(\mathbf{r},\omega) = \frac{1}{c}\sum_\lambda \left\{ F_{\lambda g}(\omega)\left(\frac{1}{E_{\lambda g} - \hbar\omega - i\gamma} - \frac{1}{E_{\lambda g}}\right)\mathbf{I}_{g\lambda}(\mathbf{r}) + F_{g\lambda}(\omega)\left(\frac{1}{E_{\lambda g} + \hbar\omega + i\gamma} - \frac{1}{E_{\lambda g}}\right)\mathbf{I}_{\lambda g}(\mathbf{r})\right\} , \tag{3.5}$$

and

$$F_{\mu\nu}(\omega) = F^{(0)}_{\mu\nu}(\omega) + \sum_\lambda [A_{\mu\nu,g\lambda}F_{\lambda g}(\omega) - A_{\mu\nu,\lambda g}F_{g\lambda}(\omega)] \ , \tag{3.6}$$

where either μ or ν is g because only $F_{\lambda g}$ and $F_{g\lambda}$ appear in the expression of $\tilde{\mathbf{j}}$, Eq. (3.5). If the electromagnetic field is resonant with excitation energies of the matter system, only the first term in $\tilde{\mathbf{j}}$ is dominant. When we take ω to 0, however, the nonresonant terms have significant contribution. To see this, we have to keep in mind that F's include $\mathbf{A}$ linearly and $\mathbf{A}$ is proportional to $\mathbf{E}/\omega$. If we neglect the nonresonant terms inversely proportional to $E_{\lambda g}$ in $\tilde{\mathbf{j}}$, it would diverge as $\omega \to 0$ for a finite electric field even if the matter system has a finite excitation energy. It is the nonresonant terms which suppress this unphysical divergence. Moreover, it is easily shown, by using the identity, (2.17), that the leading term of $\tilde{\mathbf{j}}$ is in proportion to ω at $\omega \sim 0$. This leads to the fact that the radiated electromagnetic field intensity is proportional to ω^4 at infinity, where only the dipole component contributes. This is the well known ω dependence of Rayleigh scattering.

In numerical application, we can deal with only a limited number of states. To supplement the contribution of the neglected states, it is widely accepted to introduce the background susceptibilities which are usually local. It is sometimes useful to see that the separability nature is preserved even the local susceptibility functions are introduced.[6] To this end, we write the the background current density in the following form in terms of local susceptibility tensors, as

$$\tilde{\mathbf{j}}_b(\mathbf{r},\omega) = \sum_{\xi\eta}\int d\mathbf{r}' \chi^{(b1)}_{\xi\eta}\delta(\mathbf{r}-\mathbf{r}')\tilde{\theta}(\mathbf{r})\tilde{\theta}(\mathbf{r}')\hat{\mathbf{e}}_\xi\hat{\mathbf{e}}_\eta\cdot\tilde{\mathbf{A}}(\mathbf{r}',\omega) + \sum_{\omega_1+\omega_2=\omega}\sum_{\xi\eta\zeta}\int d\mathbf{r}_1\int d\mathbf{r}_2 \chi^{(b2)}_{\xi\eta\zeta}\delta(\mathbf{r}-\mathbf{r}_1)\delta(\mathbf{r}-\mathbf{r}_2)\tilde{\theta}(\mathbf{r})\tilde{\theta}(\mathbf{r}_1)\tilde{\theta}(\mathbf{r}_2)\hat{\mathbf{e}}_\xi\hat{\mathbf{e}}_\eta\cdot\tilde{\mathbf{A}}(\mathbf{r}_1,\omega_1)\hat{\mathbf{e}}_\zeta\cdot\tilde{\mathbf{A}}(\mathbf{r}_2,\omega_2) + \cdots , \tag{3.7}$$

where $\hat{\mathbf{e}}_\xi$ is the unit vector along ξ axis ($\xi = x, y, z$) and $\tilde{\theta}$ is unity in the region which the matter fills and is zero elsewhere. By expanding the δ function in terms of a complete set $\{\varphi_i\}$ as $\delta(\mathbf{r}-\mathbf{r}') = \sum_i \varphi_i(\mathbf{r})\varphi_i(\mathbf{r}')$, the spatial integration in $\tilde{\mathbf{j}}_b(\mathbf{r},\omega)$ can be carried out and we have

$$\tilde{\mathbf{j}}_b(\mathbf{r},\omega) = \sum_{i\xi}\chi^{(b1)}_{\xi\eta}\hat{\mathbf{e}}_\xi\tilde{\varphi}_i(\mathbf{r})G_{i\eta}(\omega) + \sum_{\omega_1+\omega_2=\omega}\sum_{ij\xi\eta\zeta}\chi^{(b2)}_{\xi\eta\zeta}(\mathbf{r})\hat{\mathbf{e}}_\xi\tilde{\varphi}^{(2)}_{ij}(\mathbf{r}) \times G_{i\eta}(\omega_1)G_{j\zeta}(\omega_2) + \cdots , \tag{3.8}$$

where

$$\tilde{\varphi}_i(\mathbf{r}) = \tilde{\theta}(\mathbf{r})\varphi_i(\mathbf{r}) \ , \tag{3.9}$$

$$\tilde{\varphi}^{(2)}_{ij}(\mathbf{r}) = \tilde{\theta}(\mathbf{r})\varphi_i(\mathbf{r})\varphi_j(\mathbf{r}) \ , \tag{3.10}$$

and

$$G_{i\xi}(\omega) = \int d\mathbf{r}\tilde{\varphi}_i(\mathbf{r})\hat{\mathbf{e}}_\xi\cdot\tilde{\mathbf{A}}(\mathbf{r},\omega) \ . \tag{3.11}$$

Thus, the separability holds for the background (local) susceptibility, too. The coefficients G's are determined by a set of polynomial equations in which F's and G's are coupled. Extension to the general orders would be obvious.

Before the emergence of mesoscopic systems, the problem of nonlocal response was a rather special subject, though it was recognized to be a very fundamental one. It was discussed in connection with the spatial dispersion effect of excitons[7] and electrons in metals.[8] Both systems are characterized by the wave number dependent eigenenergies (and hence dielectric function), which inevitably leads to the existence of two (or more) coupled modes of EM wave and matter polarization for a given frequency. This gave rise to the problem of additional boundary condition (ABC),[9] which has been discussed for a long time. The very existence of the ABC problem seems to be connected with the attitude of keeping the traditional framework of optical response theory, which deals with the macroscopic electromagnetic field together with the (bulk) dielectric function of matter. The correct answer to the ABC problem was the use of the dielectric function in the site representation, with explicit information of material boundary in itself.[10] Among the proposed forms of such dielectric functions, some have separable

forms.[11] But the main stress of such models was on the possibility of carrying out the infinite summation over the wave numbers, which gives some tractable forms of integral kernels. By using such susceptibilities, one could derive explicit forms of ABC, which depend on the starting models.[11,12]

The positive use of the separability has lead to an ABC-free theory of exciton polaritons,[13,14] though the separable nature of (linear) susceptibility was model dependent at that time. The awareness of the general character of the separability has led us to the nonlocal response theory,[2] which extended the validity range of the ABC-free theory to the arbitrary shape and size of a matter and to the microscopic treatment of EM field. However, the susceptibility functions considered there were the resonant components alone, which arise from Eq. (2.23), i.e., one of the three possible origins. The present result removes this restriction, and allows us to treat all the components of linear and nonlinear susceptibilities as separable integral kernels without depending on the models of samples. In this way, the present argument contributes to the complete formulation of the nonlocal response theory.

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