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Nonlocal Response: An Unconventional Aspect of the Optical Properties of Mesoscopic Systems

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Abstract
Qualitative aspects of microscopic nonlocal theory are discussed from the viewpoint that this unconventional way of description is a very fundamental one with wide applicability to various opitical processes in condensed matter, especially in mesoscopic systems. Discussions are given on the nonlocality, the choice of Matter Hamiltonian and the gauge of EM field, an extended picture of Lorentz model, the problem of boundary conditions, background dielectric constant, the self-sustaining modes, microscopic internal field, near-field problems, and radiative and nonradiative widths.

Keyword:
nonlocal response, microscopic variation, self-sustaining modes, radiative width

INTRODUCTION

Conventional theory of the optical response of solids is based on the macroscopic Maxwell equations for electromagnetic (EM) field with an auxiliary condition, called constitutive equation. All the field variables in the equations are regarded as macroscopically averaged quantities. Namely, they are slowly-varying functions in space. In accordance with this macroscopic nature of the field variables, the constitutive equation is usually assumed to have local relationship between polarization $\boldsymbol{P}$ and electric field $\boldsymbol{E}$. In the case of linear response, it has the following form:

$$\boldsymbol{P}(\boldsymbol{r},\omega) = \chi^{(1)}(\omega)\boldsymbol{E}(\boldsymbol{r},\omega) \tag{1}$$

where the susceptibility is assumed to be a material constant, which may be frequency (ω) dependent. However, if one calculates the susceptibility quantum mechanically, the relationship turns out to be nonlocal, namely,

$$\boldsymbol{P}(\boldsymbol{r},\omega) = \int d\boldsymbol{r}' \bar{\chi}^{(1)}(\boldsymbol{r},\boldsymbol{r}';\omega)\boldsymbol{E}(\boldsymbol{r}',\omega) \tag{2}$$

where $\bar{\chi}^{(1)}(\boldsymbol{r},\boldsymbol{r}';\omega)$ is generally non-zero for $\boldsymbol{r} \neq \boldsymbol{r}'$ in a matter. This nonlocality holds for both linear and nonlinear susceptibilities.

Although the nonlocal nature of susceptibility is known in principle, our practical knowledge of optical response of condensed matter is so much dominated by the traditional way of thinking, i.e., the macroscopic local response theory, which has been very successful in the past. The appearance of mesoscopic systems has lead us to realize the limit of applicability of the macroscopic approach, and it reminds us of the microscopic nonlocal response as a more fundamental approach, although it was rather an unconventional method until recently, except for the case of additional boundary condition for excitons and plasmons [1,2]. In this paper, we will discuss various characteristic features of nonlocal response, emphasizing its general character in comparison with the macroscopic local response.

MATTER AND EM FIELD

As a first-principles theory, it is important to define matter and EM field precisely. The main choice is whether we regard the longitudinal electric field as the internal field of a matter or not. The consequence of this choice affects the form of the matter Hamiltonian.

From the solution of the microscopic Maxwell equations with a polarization $\boldsymbol{P}(\boldsymbol{r},\omega)$ as a source term, the longitudinal part of electric field is obtained generally as

$$\boldsymbol{E}_{\text{long}}(\boldsymbol{r},\omega) = \nabla\nabla \cdot \int d\boldsymbol{r}' \frac{\boldsymbol{P}(\boldsymbol{r}',\omega)}{|\boldsymbol{r}-\boldsymbol{r}'|} \,. \tag{3}$$

This means that the longitudinal field is the instantaneous Coulomb field due to the polarization charge density $-\nabla \cdot \boldsymbol{P}$. Thus, the interaction of this field with a charge in the matter is simply interpreted as Coulomb interaction between charges.

From the above argument, we are lead to a choice, which is conceptually simple. Namely, we take $\boldsymbol{E}_{\text{long}}$ (times a charge) as an internal force of the matter, regarding only the transverse part of electric field as EM (radiation) field. In this case, the matter Hamiltonian contains the full Coulomb interaction among all the charged particles.

In terms of vector and scalar potential, this is equivalent to the following : we take Coulomb gauge ($\nabla \cdot \boldsymbol{A} = 0$), and the integrated scalar potential over all space, which is equivalent to the instantaneous Coulomb potential among all the charged particles [3], is considered as a part of matter Hamiltonian. The field degrees of freedom consist of the transverse components $\boldsymbol{A}(\boldsymbol{r}, \omega)$ alone, and the matter Hamiltonian is the sum of kinetic energies and (full) Coulomb potentials.

There is an another, more frequently made choice, where $\boldsymbol{E}_{\text{long}}$ is also considered as a part of EM field acting on the matter. In this case, it is obvious that we cannot maintain the full Coulomb interaction in the matter Hamiltonian. In order to be consistent, we have to omit a certain part of Coulomb interaction from the matter Hamiltonian [4]. Such part of Coulomb interaction should be chosen as

$$\Delta H_C = -\int d\boldsymbol{r} \boldsymbol{E}_{long}(\boldsymbol{r}, \omega) \cdot \boldsymbol{P}(\boldsymbol{r}, \omega) \tag{4}$$

$$= \int d\boldsymbol{r} \int d\boldsymbol{r}' \frac{\nabla \cdot \boldsymbol{P}(\boldsymbol{r}, t) \nabla' \cdot \boldsymbol{P}(\boldsymbol{r}', t)}{|\boldsymbol{r} - \boldsymbol{r}'|} , \tag{5}$$

where the Coulomb interaction between polarization charges is rewritten as the interaction energy between the longitudinal field and the polarization.

The problem of choosing external field and corresponding matter Hamiltonian was studied in the case of excitons in 3D crystals [5], and the relevant part of the matter Hamiltonian to be subtracted was shown to be the electron-hole exchange interaction H_{exch}. (Note that ΔH_C is equivalent to the electron-hole exchange interaction [6].) However, the above argument seems to support the answer in more general situations than 3D crystals.

In the case of bulk material, H_{exch} causes the energy splitting between the transverse and longitudinal polarization waves of excitons and optical phonons. This splitting may be described as the energy difference between matter eigenstates, or as due to the difference in the coupling with depolarization field.

EXTENDED LORENTZ PICTURE

Once we choose the Coulomb gauge and the matter Hamiltonian containing the full Coulomb interaction among the charged particles, we are ready to start the selfconsistent determination of the microscopic motions of the matter and EM field. As mentioned in the introduction, the macroscopic response theory needs to be modified, so that it includes the nonlocal character arising from the microscopic quantum mechanical calculation.

The details of such a framework will be left for references [7-9], but we will describe the physical picture of this approach in this note.

In the Maxwell equations the matter system is represented by current and charge densities and the current density may be considered as an independent quantity because of the continuity equation. This quantity consists of various components of characteristic frequencies corresponding to the transition energies of the matter system. In the absence of (transverse) EM field, they are just independent oscillators, possibly with damping effect. In the presence of EM field there are two effects on the current density: The oscillation is induced for each of its components, the amplitude of which depends on the frequency detuning, the coupling strength and damping. The second effect is the exchange of transverse EM field among the components of the current density.

According to the classical Lorentz model, a matter is assumed to consist of an assembly of electric oscillators. This model well describes the frequncy dependence of the optical response of a matter including resonance behavior, as the (macroscopically averaged) quantum mechanical calculation does.

It is interesting that the physical picture of interacting radiation- matter system described by the microscopic nonlocal theory may be viewed as an "extended Lorentz model". Namely, a matter is an assembly of oscillating current densities with characteristic eigenfrequencies. Though such a description is "a fact" derived from a first-principles theory rather than "a model", it is a same physical picture as the original Lorentz model.

There are two aspects generalized from the original Lorentz model: (i) Each current density has a quantum mechanical (coherent) spatial extension which can be larger than the wavelength of interacting EM field. This requires the nonlocal description of the optical response, which leads to a selfconsistent determination of the spatial variations of current density and EM field. (ii) Various current densities interact with one another via the exchange of transverse EM field.

The second aspect (ii) is an essential ingredient of the microscopic nonlocal formalism. Each component of the current density is oscillating in time, and it emits transverse EM field according to the Maxwell equations. The emitted EM field propagates with the light velocity, and interacts with another components of current densities. Such a process may be called the retarded interaction between current densities.

The interaction energy due to this process is written as the interaction of one component of current density with the transverse field produced by the other. The components of current densities are defined as the matrix

element of current density operator $\hat{\mathbf{I}}(\boldsymbol{r})$ with respect to the eigenstates ($|\mu>, |\nu>, etc.$) of the matter. The retarded interaction among them for frequency ω is given as

$$\tilde{A}_{\mu\nu,\tau\sigma} = \frac{1}{c^2} \int d\boldsymbol{r} \int d\boldsymbol{r}' < \mu|\hat{\boldsymbol{I}}(\boldsymbol{r})|\nu > \cdot \tilde{\mathbf{G}}_q(\boldsymbol{r}, \boldsymbol{r}') \cdot < \tau|\hat{\boldsymbol{I}}(\boldsymbol{r}')|\sigma > \qquad (6)$$

where $\tilde{\mathbf{G}}_q(\boldsymbol{r}, \boldsymbol{r}')$ is the radiation Green function in vacuum ($q = \omega/c$), and $\hat{\boldsymbol{I}}(\boldsymbol{r})$ is the current density operator in the absence of vector potential.

The inclusion of this effect allows us a correct description of the "size, shape, and internal structure"-dependence of optical responses with the resonances of the coupled-modes which cover various cases such as polaritons, dynamically scattered X-rays, surface modes, whispering gallery modes, radiatively decaying level of an atom, etc.

BOUNDARY CONDITIONS

In the conventional theory of optical response, a typical way of solving problems is, among others, to solve the Maxwell equations in each region of space which is described by a single dielectric constant, and to connect the solutions across the boundary of such regions. The boundary conditions to be used for such a purpose are derived from the macroscopic Maxwell equations, leading to the continuity of the tangential components of electric or magnetic field and/or the normal component of displacement field across the boundary.

This method of solution gives a complete answer as long as all the dielectrics in the system are described by local dielectric constants. When we are interested in resonant response due to excitons or plasmons, however, the relevant (bulk) dielectric function depends not only on frequency, but also on wave vector $\boldsymbol{k}$, reflecting the $\boldsymbol{k}$-dependence of the energy of the elementary excitations in question. Such a medium is called spatially dispersive, and has provided a long standing controversy as to the necessity and the actual form of the so-called additional boundary condition (ABC) since the first paper by Pekar [10].

Looking back the history of ABC problem, we can now see that it arose from the inappropriate approach to a problem of *finite* sysytem in terms of the concept for *infinite* crystals. Namely, one started from the solution of the dispersion equation $\epsilon(\boldsymbol{k}, \omega) = (ck/\omega)^2$ for a bulk crystal, two or more solutions of which for a given ω necessitates ABC (or ABC's) in order to have a unique connection to the field outside the spatially dispersive medium.

The correct approach to this problem is to start with the dielectric function or electric susceptibility which explicitly includes the effect of sample surface. Namely, we should rely on the response functions in site representation. This is essentially the approach of the microscopic nonlocal theory mentioned above. Then, we can either derive the explicit form of the ABC appropriate for adopted model [11-13], or solve the Maxwell equations without referring to any ABC [14,15].

The only consideration of boundary condition in the nonlocal theory is that we choose the outgoing solution of the Maxwell equations, i.e., the EM field originating from the source polarization should go away from the source with time. But this is of course different from the boundary condition discussed above.

The problem of boundary condition (including ABC) exists only in the macroscopic treatments of the radiation-matter interaction. However, a mixed situation may arise, as discussed in the next subsection.

BACKGROUND DIELECTRIC CONSTANT

In a truely microscopic treatment, all the excited levels of matter should be described quantum mechanically. However, it is not realistic to treat an infinite number of degrees of freedom. Neither is it physically effective. In most cases of microscopic treatment, we are interested in some resonant processes. Therefore, a convenient and often suscessful method is to treat a few resonant levels quantum mechanically, and regard the rest of the exicted levels as the source of background dielectric constant, which is usually considered to represent a local medium.

Since a local medium is familiar in macroscopic response, the above situation can be treated as a mixed problem of micro- and macroscopic response. Since the macroscopic part of polarization is described by local susceptibility, which does not contain any information of the size and shape of sample, we need the boundary conditions (in usual sense) to fix the response EM field. If the resonant part of polarization is treated by the nonlocal formalism, there is no necessity of ABC [14,15].

However, even such a mixed situation could be handled by the nonlocal formalism, if we rewrite the local susceptibility in terms of a complete set of functions $\{\phi_\nu\}$ as

$$\chi(\omega)\delta(\boldsymbol{r}-\boldsymbol{r}') = \chi(\omega)\sum_\nu \phi_\nu(\boldsymbol{r})^*\phi_\nu(\boldsymbol{r}') \ . \tag{7}$$

Because of the separable form of this quantity as an integral kernel, this fits well to the framework of the nonlocal theory [16], which is free from any

boundary conditions. In numerical calculation one has to limit the number of $\{\nu's\}$. This may spoil the exactness of the above expansion. However, it will not seriously affect the response of the resonant level(s) in question, because the main characteristics of response spectrum is determined by the resonant part of polarization.

The following comment will be worth mentioning. In the second subsection, a discussion has been given as to the two possibilities of choosing the EM field acting on a matter. In one case, it is the complete Maxwell field containing the transverse and longitudinal parts, and in the other, it is only the transverse part. Both of them are correct if the coresponding Hamiltonian of matter is properly chosen. Two different susceptibilities are defined to describe the same polarization. The relationship between them can be explicitly given in the case of infinite 3D crystals [17]. When we want to separate the susceptibility into resonant and nonresonant parts, and regard the latter as a local background (constant) susceptibility, it is appropriate to do so only for the susceptibility defined with respect to the whole Maxwell field. Because, in the other case, the matter Hamiltonian contains the self-interaction of induced polarization, even the background part of susceptibility should depend on the shape and size of sample, which cannot be written in the usual form of background susceptibility.

SELF-SUSTAINING MODES

In a semiclassical theory of radiation-matter interaction, one looks for the selfconsistent motion of EM field and induced current density (or polarization) for a given initial condition such as an incident EM field. This requirement leads to a coupled integral equations for vector potential $\boldsymbol{A}(\boldsymbol{r},\omega)$ and current density $\boldsymbol{j}(\boldsymbol{r},\omega)$. In the microscopic nonlocal formalism, these coupled equations are shown to further reduce to a coupled algebraic equations for the new unknowns

$$F_{\mu\nu}(\omega) = \int d\boldsymbol{r} < \mu|\hat{\boldsymbol{I}}(\boldsymbol{r})|\nu > \cdot \boldsymbol{A}(\boldsymbol{r},\omega) \ . \tag{8}$$

In the case of linear response, the coupled equations are a set of linear equations of the (matrix) form $\mathbf{SF} = \mathbf{F^{(0)}}$, where $F^{(0)}_{\mu\nu}$ is defined by the same equation as $F_{\mu\nu}$ with $\boldsymbol{A}$ replaced by the incident field $\boldsymbol{A}_0$. The coefficient matrix $\mathbf{S}$ contains the transition energies of the matter and the retarded interaction, eq.(6), among induced current densities. Both the response field and current density are linear functions of $F_{\mu\nu}$ with known coefficients. Thus the solution $\mathbf{F} = \mathbf{S}^{-1}\mathbf{F^{(0)}}$ completely determines the

response field and induced current density at any point $\boldsymbol{r}$ and frequency ω. [1]

The condition $det(\mathbf{S}) = 0$ gives the finite amplitude solution in the absence of the incident field. Since the induced current density and EM field are supporting one another to make up this solution, it may be called "self-sustaining (SS) mode". An important general aspect is that its eigen frequency, generally a complex number, appears as a pole of the response ($\propto \mathbf{S}^{-1}$). The value of this comlpex pole represents an excitation energy of matter with the correction of radiative shift and width. Therefore, resonant peaks in response spectra do not simply give the values of matter excitation energies. In mesoscopic systems the amount of the radiative correction is not at all negligible compared with the spacings between the matter excitations, which could lead to an interesting novel effect [18].

The physical picture of the self-sustaining modes varies from system to system quite remarkably: (a) The SS mode of an infinite 3D crystal in the energy region of phonons (excitons) is phonon- (exciton-) polaritons. It has no radiative decay, because there is no outer space for photons to escape to. The SS mode in X-ray region is the multiply scattered X-rays satisfying the dispersion relation of dynamical scattering regime, which has tiny forbidden gaps in certain directions of the crystal [19]. One can also generalize the description in terms of SS modes to include "polaritons with lattice diffraction effect", "X-ray dynamical scattering in the presence of resonant transitions", or "the photonic bands" of a periodic array of dielectrics [17]. (b) The SS modes of a crystal surface are surface exciton (plasmon, phonon, etc.) polaritons. (c) The SS modes of a sphere are Mie resonances, and whispering gallery modes in the case of a nonmetallic sphere with radius larger than light wavelength. (d) In various types of cavity including a microcavity, the SS modes are the cavity modes when it is empty. When it contains a matter with resonant levels, the SS modes are the coupled cavity-matter resonances, which may show vacuum Rabi splittings. This coupled mode is sometimes called cavity polariton [20,21], and is claimed to be a new and essential basis to discuss nonlinear optical effects. However, this is just an example of the general picture of SS modes which contribute to the resonances of optical spectra. (e) In the case of an isolated atom in vacuum, its SS mode is an atomic excitation with radiative shift and width. The radiative width of the ν-th level, i.e., the imaginary part of $\tilde{A}_{0\nu,\nu 0}$ of (6), is exactly the same expression obtained from a QED calculation [22].

[1] For nonlinear processes, the matrix $\mathbf{S}$ contains also the components of $\mathbf{F}$, and the number of the unknowns is increased with more μ and ν and the frequency components.

MICROSCOPIC INTERNAL FIELD

It should be stressed that the EM field and induced current density obtained from the microscopic nonlocal theory have microscopic spatial variations corresponding to the quantum mechanical description of nonlocal susceptibilities. In micro- and mesoscopic systems, the quantized levels are well separated, so that each resonance with one of these levels is characterized, not only by its energy position, but also by its spatial structure of induced current density. Suppose the wavelength of the resonant light is λ_0 in vacuum. Then, it should be noted that the spatial structure of resonantly induced current density can have much shorter wavelength component than λ_0, and that the corresponding spatial variation should appear also in the selfconsistently determined EM field (internal field) [23].

Such a spatial structure reflects the matrix element of the current density for the transition, and thus it is quite different for each resonance. The spatial structure of EM field for a given frequency consists of resonant and nonresonant contributions. The resonant part is, as mentioned above, reflects the properties of the resonant level sensitively, while the nonresonant part, arising from all the other nonresonant levels, shows only gradual change with frequency.

The ratio of the resonant and nonresonant contributions is sensitive to the magnitude of the transition dipole moment and the non-radiative width of the resonant level [18].

Within linear response scheme, the physical picture of the spatial variation of the internal field is just a different way of describing the light interference in a confined matter system. However, in nonlinear response, this way of describing the internal EM field brings about an additional point of view in picking out important processes. Namely, a large nonlinear signal arises, not only from energetically resonant processes, but also from those involving large amplitudes of internal field.

In a pump-probe process with different frequncies, for example, a resonant light may produce a large amplitude of EM field with a particular spatial structure. In its presence, there can be a transition between matter excited states that contributes to the nonlinear susceptibility. If the second light is made resonant with this transition, we can expect a large signal of the pump-probe process [18,24].

In iterative treatments of nonlinear processes, the field acting on matter is an incident one and those produced iteratively [25]. This will be acceptable for nonresonant processes. However, the nonlocal framework, being most valuable for resonant processes, requires, not iterative, but a self-consistent treatment of field and current density. This produces a

resonant component of EM field with a rapid spatial variation (compared with that of incident light), which is absent in iterative treatment.

NEAR-FIELD PROBLEMS

Near-field spectroscopy or scanning near-field optical microscopy (SNOM) is also an appropriate problem for the microscopic nonlocal response theory, because the near-field of a quantum mechanical object can be discussed only by a microscopic treatment. But the degree of proximity in talking about "near"-field is diverse, depending of the problems in consideration. For non-resonant processes, matter system may be described by local dielectric constants, and thus the degree of "nearness" is limited by the macroscopic nature of the framework. This type of system will be of interest from applicational viewpoint, but less challenging from the fundamental one for theory, because the problem is just to solve the macroscopic Maxwell equations with complicated boundary conditions. The more challenging problems would be those related with the near-field in microscopic scale.

A particularly interesting case for theory would be the near-field produced by a resonant light. Such a field has a microscopic structure reflecting the nature of the relevant wave functions. With a high-resolution probe-tip one should in priciple be able to detect the microscopic position dependence and the frequency dependence of the near-field intensity. In this way, a resonant SNOM provides a new type of spectroscopy producing the $(\boldsymbol{r},\omega)$-map of the near-field or induced polarization on the sample.

The signal map can also be much dependent on the ways to excite sample and detect signal. The dependence of signal map on the operation mode (irradiation, reflection, and illumination modes) of resonant SNOM is going to be discussed in this conference [26]. There is a remarkable difference in signal by changing the mode of operation.

A peculiar effect in resonant SNOM is configuration resonance [27]. This is due to the change in the resonant frequency of the whole system as probe-tip is scanned. Since the scanning is usually done with a fixed frequency of observation, there occurs a position dependent detuning. This causes a strong variation on the amplitude of induced polarization on sample and probe-tip, which is reflected on the intensity distribution of the signal. Though the effect may sometimes increases the spatial resolution, it has also an effect of distorting the sample image [28]. Therefore, one should be careful in interpreting the signal.

Another peculiarity in near-field spectroscopy is that the field for excitation and detection may not be spatially slowly-varying. This suggests

that the optical selection rule based on the multipole expansion of EM field, which is a standard key concept in macroscopic response theory, may become useless. A typical case would be the reflection mode SNOM, which excites the sample and detects the signal through the microscopic probe-tip. In this case, the EM field cannot be treated in long wavelength approximation, and therefore there is no basis to talk about the electric dipole selection rule. A model calculation was made with a simple 1D chain of fine particles, and it was demonstrated that both electric-dipole allowed and forbidden transitions contribute to the signals with almost same intensity [29].

RADIATIVE AND NONRADIATIVE WIDTHS

Finally, a short comment will be given about the different origins of the widths of spectral peaks. Such a width is always related with an isolated level coupled with continuous levels. According to the origin of the latter, the width is called radiative or non-radiative. For EM field, the existence of a continuum is rather obvious. A high-Q cavity may seem to have isolated levels, but a slight coupling with the continuum outside the cavity produces the high but finite Q-factor. The matter levels can have finite widths for various reasons such as phonons, electron-electron interactions, etc. It is necessary that the degrees of freedom can be regarded very large. In the limit of an atom or simple molecule, it is not possible to talk about nonradiative widths. It might be interesting to study at which size the concept of nonradiative width becomes valid as we increase the size of matter starting from an atom.

It should be noted that the radiative width is not included in the susceptibility function, but that it appears in the response spectrum after solving the self-consistent equations for EM field and matter, $\mathbf{SF} = \mathbf{F}^{(0)}$. The iterative treatment of these coupled equations does not give any radiative width. It is sometimes assumed to plug "a radiative width" in the susceptibility function. This is of course not correct, because, if one uses such a susceptibility to get a self-consistent solution of the radiation-matter system, it would lead to a double counting of radiative width.

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