32.1 Introduction

You might think that the great success of quantum electrodynamics (QED) would settle the debate on the nature of light to provide a clear view of its behavior, where the photon is regarded as the unit of excitation associated with a quantized mode of the electromagnetic (radiation) field. However, Heisenberg’s uncertainty principle tells us that a state of definite momentum, energy, and polarization associated with a plane wave used as a basis function of quantization must be completely indefinite in space and time. It suggests the difficulty of spatial localization of a photon as a particle. In fact, Newton and Wigner showed that a free photon, as a massless particle with spin 1, has no localized states on the basis of natural invariance requirements that localized states for which operators of the Lorentz group apply should be orthogonal to the undisplaced localized states, after a translation (Newton and Wigner 1949). According to them, one can obtain a general expression for a position operator for massive particles and for massless particles of spin 0 or 1/2, not for massless particles with finite spin, which indicates that there is no probability density for the position of the photon, and thus a position-representation wave function cannot be consistently introduced. It has also been shown that photons are not localizable, on the basis of im primitive representations of the Euclidean group (Wightman 1962). It is now believed that photons are only weakly localizable, although single-photon states with arbitrarily fast asymptotic falloff of energy density exist, and that a lack of strict localizability is directly related to the absence of a position operator for a photon in free space and a position-representation photon wave function (Hawton 1999).

On the other hand, several authors have claimed that a minimum modification of the naive route leads to a wave-function description of a photon, even though the probability density for the position of a photon and a position-representation wave function cannot be consistently introduced. For example, it has been shown to be possible to introduce a position-representation wave function \( \psi(\vec{r}, t) \) for a photon, which is the expectation value of the photon energy in a region \( d\vec{r} \) about \( \vec{r} \) (Sipe 1995, Scully and Zubairy 1997, Hawton 1999, Roychoudhuri and Roy 2003). Mandel et al., from another viewpoint, have found that a photon wave function as a probability amplitude is possible in a coarse-grained volume whose linear dimensions are larger than the photon wavelength (Mandel 1966, Mandel and Wolf 1995, Inagaki 1998). The localization of the photon energy density and photodetection rates having an exponential or arbitrarily fast asymptotic falloff have been discussed, as well as causality (Hegerfeldt 1974, Pike and Sarkar 1987, Hellwarth and Nouchi 1996, Adlard et al. 1997, Bialynicki-Birula 1998, Keller 2005).

When the interactions with matter were considered, different views opened up. Dressed states and operators, dressed and half-dressed sources in nonrelativistic QED, electromagnetic field correlations, and intermolecular interactions between molecules in either ground or excited states have been discussed (Compagno et al. 1988, 1995, Cohen-Tannoudji et al. 1989, 1992, 1996, Adlard et al. 1997, Bialynicki-Birula 1998, Keller 2005).
Power and Thirunamachandran 1993), focusing on the fact that a bare source interacting with a quantum field is surrounded by a cloud of virtual particles. It has been shown that the dressing of the source, or virtual cloud effects, can be detected by a test body (detector) located close to the source. Carniglia and Mandel (1971) proposed a complete basis for electromagnetic fields interacting with a material of refractive index $n$ filled in a half space separated by vacuum in order to quantize the source fields, while Inoue and Hori (2001) discussed the detector modes and the behavior of a photon–atom interacting system near the material surface. Kobayashi et al. (2001) focused on the environmental effects on a nanomaterial interacting with photons and obtained a near-field optical interaction as an effective interaction between nanomaterials electronically disconnected, but closely located, in order to detect the cloud of virtual photons. They have also applied it to a discussion of nanophotonic devices (Sangue et al. 2004). Focusing on the photon degrees of freedom, on the other hand, photon hopping has been employed to discuss a photon–material interacting system (John and Quang 1995, Suzuura et al. 1996, Shojiyuchi et al. 2003), and a photon dressing by material excitations has been recently discussed using the photon-hopping model in real space and a quasiparticle model (Kobayashi et al. 2008).

Focusing on light–matter interactions at the nanoscale, we discuss a near-field optical interaction between nanomaterials surrounded by a macroscopic system, a dressing mechanism, and spatial localization of photons in this article, which is organized as follows. Section 32.2 is devoted to background issues for photon localization, in particular, the difficulty of the definition, effective methods, and free-field quantization. In Section 32.3 we discuss a dressing mechanism of photons and their localization in space, including virtual clouds of photons, electromagnetic field correlations and intermolecular interactions, effective near-field optical interactions, and phonons’ effects on photon localization. Finally, a summary and a future outlook are presented.

### 32.2 Background

#### 32.2.1 Difficulty in Defining a Wave Function of a Photon in Real Space

Since there is no probability density for the photon, and thus a position-representation wave function in free space cannot be consistently defined, one has to follow quantum electrodynamics (QED), that is, to redefine one- and a few-photon wave functions in a physically meaningful way in order to obtain fruitful insights into the photon–matter interacting system at the nanoscale. In the following subsections, we will give an overview of both approaches, after pointing out the difficulties involved with a position-representation wave function of a photon in free space.

We begin with Maxwell’s equations in vacuum:

\[ \nabla \times \vec{E}(\vec{r}, t) = \frac{1}{c} \frac{\partial \vec{B}(\vec{r}, t)}{\partial t}, \]  

(32.1a)

where

\[ c \]  

is the speed of light

\[ \vec{E}(\vec{r}, t) \]  

and \[ \vec{B}(\vec{r}, t) \] are electric and magnetic fields, respectively.

Let us now define \( \Phi_\pm(\vec{r}, t) \) as \( \Phi_\pm(\vec{r}, t) = \vec{E}(\vec{r}, t) \pm i\vec{B}(\vec{r}, t) \); then, it follows from Maxwell’s equations that \( \Phi_\pm(\vec{r}, t) \) should satisfy

\[ \frac{i\hbar}{\partial} \Phi_\pm(\vec{r}, t) = \pm \hbar \nabla \times \Phi_\pm(\vec{r}, t). \]  

(32.2)

Using the Fourier transform of these functions, we introduce the vectors \( \tilde{\gamma}_\pm(\vec{p}, t) \) as

\[ \tilde{\Phi}_\pm(\vec{r}, t) = \int \frac{d^3 \vec{p}}{(2\pi \hbar)^3} \tilde{\gamma}_\pm(\vec{p}, t) e^{i\vec{p} \cdot \vec{r}} e^{i\hbar \vec{p} \cdot \vec{r}}, \]  

(32.3)

which is associated with \( \gamma_\pm(\vec{p}, t) \) as \( \tilde{\gamma}_\pm(\vec{p}, t) = \gamma_\pm(\vec{p}, t) \tilde{e}_\pm(\vec{p}) \) with

\[ \tilde{e}_\pm(\vec{p}) = \frac{1}{\sqrt{2}} [\tilde{e}_\pm(\vec{p}) \pm \tilde{e}_\pm(\vec{p})], \]  

(32.4a)

where two unit vectors \( \tilde{e}_\pm(\vec{p}) \) and \( \tilde{e}_\pm(\vec{p}) \) are defined such that the unit vectors

\[ [\tilde{e}_\pm(\vec{p}), \tilde{e}_\pm(\vec{p}), \vec{p} = \vec{p} \parallel \vec{p} \]  

(32.4b)

form a right-handed triad. Then, it is easy to verify that \( \gamma_\pm(\vec{p}, t) \) satisfy a Schrödinger-like equation in the momentum representation

\[ \frac{i\hbar}{\partial} \gamma_\pm(\vec{p}, t) = cp \gamma_\pm(\vec{p}, t), \]  

(32.5)

which indicates that \( \gamma_\pm(\vec{p}, t) \) are probability amplitudes for photons of momentum \( \vec{p} \), energy \( E = cp \) with \( p = |\vec{p}| \), and positive/negative helicity. Here note that

\[ \tilde{\gamma}_\pm(\vec{p}, t) \cdot \tilde{\gamma}_\pm(\vec{p}, t) d\vec{p} = \gamma_\pm^*(\vec{p}, t) \cdot \gamma_\pm(\vec{p}, t) d\vec{p} \]  

(32.6a)

and

\[ \tilde{\gamma}_\pm(\vec{p}, t) \cdot \tilde{\gamma}_\pm(\vec{p}, t) = 0 \]  

(32.6b)
show the probability of detecting a photon of positive helicity and momentum $\vec{p}$ between $\vec{p}$ and $\vec{p} + d\vec{p}$, and likewise for a photon of negative helicity. Then the normalization condition is

$$\int \left[ \gamma^+_s(\vec{p}, t) \cdot \gamma_+(\vec{p}, t) + \gamma^-_s(\vec{p}, t) \cdot \gamma_-(\vec{p}, t) \right] d^3p = 1,$$  
(32.7)

which leads to

$$\int \left[ \Phi^+_s(\vec{r}, t) \cdot \Phi_+(\vec{r}, t) + \Phi^+_s(\vec{r}, t) \cdot \Phi_+(\vec{r}, t) \right] d^3r = 1,$$  
(32.8)

and it follows from the dynamical equations that

$$\Phi_s(\vec{r}, t) = \int \frac{d^3p}{(2\pi\hbar)^3} \gamma_+(\vec{p}, 0) e^{i\vec{p} \cdot \vec{r}/\hbar}.$$

Here we note that the sum of $\Phi_+(\vec{r}, t)$ might be regarded as the position-representation wave function of a photon, but it cannot be regarded as such because Newton and Wigner and also Wightman have shown that the photon, being a massless particle, is not localizable in free space, and that there does not exist a probability amplitude and density for the position of the photon in the usual sense (see also (32.15a) and (32.15b)) (Newton and Wigner 1949, Wightman 1962).

### 32.2.2 Effective Spatial Wave Function of a Single Photon

In order to avoid the difficulties mentioned above, Mandel defined an operator representing the number of photons in a volume $V$ as the integral over $V$ of a so-called “detection operator,” which led a simple formula for the probability that $n$ photons are present in $V$ when the linear dimensions of $V$ are larger than the wavelength of light used (Mandel 1966).

Sipe took another approach to this issue, seeking a probability amplitude $\Psi_s(\vec{r}, t)$ for the photon energy to be detected about $d\vec{r}$ of $\vec{r}$ (Sipe 1995). Assuming that the integral of $\Psi^*(\vec{r}, t) \cdot \Psi(\vec{r}, t) d^3r$ over all space is proportional to the photon energy, we normalize it as follows:

$$\int \Psi^*(\vec{r}, t) \cdot \Psi(\vec{r}, t) d^3r = \int \frac{d^3p}{(2\pi\hbar)^3} \gamma_+(\vec{p}, 0) e^{i\vec{p} \cdot \vec{r}/\hbar}.$$

It is easily shown that if $\Psi_s(\vec{r}, t)$ is set as

$$\Psi(\vec{r}, t) = \Psi_-(\vec{r}, t) + \Psi_+(\vec{r}, t),$$

(32.11a)

the normalization condition is satisfied. Here note that

$$\gamma^+_s(\vec{p}, t) \cdot \gamma_+(\vec{p}, t) = 0 = \gamma^+_s(\vec{p}, t) \cdot \gamma_-(\vec{p}, t)$$

(32.12)

and

$$i\hbar \frac{\partial \Psi_s(\vec{r}, t)}{\partial t} = \pm c \hbar \nabla \times \Psi_s(\vec{r}, t).$$

(32.13)

At the same time, $\Psi_s(\vec{r}, t)$ should be chosen to satisfy an initial condition given by

$$\Psi_s(\vec{r}, t) = \frac{1}{(2\pi\hbar)^3} \int \frac{d^3p}{(2\pi\hbar)^3} \gamma_+(\vec{p}, 0) e^{i\vec{p} \cdot \vec{r}/\hbar}.$$

(32.14)

Since $\gamma^+_s(\vec{p}, t)$ and $\Psi_s(\vec{p}, \vec{r})$ are not the Fourier transform pairs, the arguments of the photon momentum $\vec{p}$ and the position $\vec{r}$ associated with the photon energy are not conjugate variables. When $\Phi_s(\vec{r}, t)$ and $\Psi_s(\vec{r}, t)$ are related by a local kernel, it can usually be regarded as a particle. However, $\Phi_s(\vec{r}, t)$, the Fourier transform of $\gamma^+_s(\vec{p}, t)$, is not a reasonable candidate for the position-representation wave function of the photon because of the following relation between $\Phi_+(\vec{r}, t)$ and $\Psi_s(\vec{r}, t)$

$$\Phi_s(\vec{r}, t) = \int w(\vec{r} - \vec{r}') \Psi_s(\vec{r}', t) d^3r'$$

(32.15a)

with the nonlocal kernel

$$w(\vec{r} - \vec{r}') = \frac{1}{(2\pi\hbar)^3} \frac{e^{i\vec{k} \cdot (\vec{r} - \vec{r}')}}{\sqrt{\hbar c k}} d^3k.$$

(32.15b)

Nevertheless, $\Psi_s(\vec{r}, t)$ might be meaningful to describe the dynamics of a photon such as a spontaneous emission from an atom and the inverse process, or at least we can detect a photon, within the range of a detector’s precision by placing a detector like an atom close to the source. In other words, it indicates that light-matter interactions near the source play an important role and should be treated consistently.

### 32.2.3 Canonical Field Quantization: Mode Functions, Field Operators, and Quantum States

It is natural to follow the canonical quantization of the electromagnetic field as a starting point for a discussion of light-matter interactions at the nanoscale. Since a lot of famous textbooks on
QED or quantum field theory have been published, we follow the essence of the theory and restrict ourselves to the free field that is free from charges and currents and whose scalar potential can be set to zero (Sakurai 1967, Roychoudhuri and Roy 2003). In the Coulomb gauge with \( \nabla \cdot \vec{A}(\vec{r}, t) = 0 \) for the vector potential \( \vec{A}(\vec{r}, t) \), three basic equations we work with for the free-field case are

\[ \vec{B}(\vec{r}, t) = \nabla \times \vec{A}(\vec{r}, t), \quad (32.16a) \]

\[ \vec{E}(\vec{r}, t) = \frac{1}{c} \frac{\partial \vec{A}(\vec{r}, t)}{\partial t}, \quad (32.16b) \]

\[ \nabla^2 \vec{A}(\vec{r}, t) - \frac{1}{c^2} \frac{\partial^2 \vec{A}(\vec{r}, t)}{\partial t^2} = 0. \quad (32.16c) \]

We expand \( \vec{A}(\vec{r}, t) \) into a complete set of mode functions \( \vec{u}_{\ell, \alpha}(\vec{r}) \) defined by the Helmholtz equation

\[ (\nabla^2 + k^2)\vec{u}_{\ell, \alpha}(\vec{r}) = 0, \quad (32.17) \]

and the boundary conditions set by the shape of a virtual cavity, for example, a box taken to be a cube of side \( L = V^{1/3} \). It follows from the Coulomb gauge that the direction of \( \vec{u}_{\ell, \alpha}(\vec{r}) \) has to be orthogonal to the wave vector \( \vec{k} \), and thus there are two polarization degrees of freedom indicated by the index \( \alpha \). Note that the mode functions become more complicated, or even impossible for more sophisticated cavity shapes. The mode functions satisfy the orthonormality condition

\[ \frac{1}{V} \int d^3r \vec{u}_{\ell, \alpha}^*(\vec{r}) \cdot \vec{u}_{\ell', \alpha'}(\vec{r}) = \delta_{\ell \ell'} \delta_{\alpha \alpha'}, \quad (32.18) \]

where we have for simplicity combined the three components of the wave vector \( \vec{k} \) and the polarization index \( \alpha \) to one index \( \ell \). Using the normalization constant \( k_l \) and the time-dependent amplitude \( q_l(t) \), we have

\[ \vec{A}(\vec{r}, t) = \sum_{\ell} \left[ A_\ell q_l(t) \vec{u}_{\ell, \alpha}(\vec{r}) + A_\ell^* q_l^*(t) \vec{u}_{\ell, \alpha}^*(\vec{r}) \right], \quad (32.19) \]

where \( q_l(t) \) follows from the differential equation of a harmonic oscillator of frequency \( \Omega_l = c|\vec{k}_l| \):

\[ \frac{d^2 q_l(t)}{dt^2} + \Omega_l^2 q_l(t) = 0. \quad (32.20) \]

The Hamiltonian of the field is

\[ H = \frac{1}{8\pi} \int d^3r (\vec{E}^2 + \vec{B}^2) = \frac{1}{8\pi} \int d^3r \left[ \left( \frac{1}{c} \frac{\partial \vec{A}(\vec{r}, t)}{\partial t} \right)^2 + (\nabla \times \vec{A})^2 \right], \quad (32.21) \]

Noticing a typical term for the \( \vec{E}^2 \) integration

\[ \int \left( \frac{1}{c} \frac{dq_l}{dt} \right) \left( \frac{1}{c} \frac{dq_{l'}}{dt} \right) \vec{u}_l(\vec{r}) \cdot \vec{u}_{l'}(\vec{r}) d^3r = \left( \frac{\Omega_l}{c} \right) V |q_l|^2 \delta_{ll'} \]  
\[ \quad (32.22) \]

and for the \( \vec{B}^2 \) integration

\[ \int (\nabla \times \vec{u}_l)(\nabla \times \vec{u}_{l'}^*) d^3r = \int (\nabla \cdot (\vec{u}_l \times \vec{u}_{l'}^*)) d^3r + \int \vec{u}_l \cdot \nabla \times (\nabla \times \vec{u}_{l'}^*) d^3r = \int \vec{u}_l \cdot (\nabla (\vec{u}_{l'}^*)) d^3r = - \int \vec{u}_l \cdot \nabla \vec{u}_{l'}^* d^3r \]

\[ = \left( \frac{\Omega_l}{c} \right)^2 V \delta_{ll'} \quad (32.23) \]

we obtain

\[ H = \frac{V}{2\pi} \sum_{\ell} \left( \frac{\Omega_l}{c} \right)^2 (A_\ell q_l^*) (A_\ell q_l). \quad (32.24) \]

If we define

\[ Q_l = \frac{1}{c} (q_l + q_l^*), \quad (32.25a) \]

\[ P_l = -i \frac{\Omega_l}{c} (q_l - q_l^*), \quad A_\ell = \frac{\sqrt{4\pi}}{V}, \quad (32.25b) \]

the Hamiltonian can be expressed in terms of a collection of independent and uncoupled harmonic oscillators as

\[ H = \sum_{\ell} \frac{1}{2} (P_l^2 + \Omega_l^2 Q_l^2). \quad (32.26) \]

Here, \( P_l \) and \( Q_l \) are seen to be canonical variables:

\[ \frac{dP_l}{dt} = \frac{\partial H}{\partial Q_l}, \quad (32.27a) \]

\[ \frac{dQ_l}{dt} = \frac{\partial H}{\partial P_l}. \quad (32.27b) \]

The natural method to quantize the field is to replace the variable \( q_l \) and its conjugate momentum \( p_l = dq_l/dt \) by operators \( \hat{q}_l \) and \( \hat{p}_l \) that satisfy the commutation relations \( [\hat{q}_l, \hat{p}_l] = i\hbar \delta_{ll'} \), or \( [Q_l, P_l] = i\hbar \delta_{ll'} \). Then, the Hamiltonian operator for the quantized field can be written as follows:
Photon Localization at the Nanoscale

\[ \hat{H} = \sum_{\ell} \frac{1}{2} (\hat{P}_i^2 + \Omega_i^2 \hat{Q}_i^2). \]  

(32.28)

Next we consider linear combinations of \( \hat{P}_i \) and \( \hat{Q}_i \) given by

\[ \hat{a}_i = \sqrt{\frac{\Omega_i}{2\hbar}} (\hat{Q}_i + i \frac{\hbar}{\Omega_i} \hat{P}_i), \]  

(32.29a)

\[ \hat{a}_i^\dagger = \sqrt{\frac{\Omega_i}{2\hbar}} (\hat{Q}_i - i \frac{\hbar}{\Omega_i} \hat{P}_i), \]  

(32.29b)

where we insert a factor to make \( \hat{a}_i \) and \( \hat{a}_i^\dagger \) dimensionless and thus satisfy the following boson commutation relations:

\[ [\hat{a}_i, \hat{a}_c^\dagger] = \frac{i}{2\hbar} [\hat{Q}_i, \hat{P}_c] + \frac{i}{2\hbar} [\hat{P}_i, \hat{Q}_c] = \delta_{ic}. \]  

(32.30)

Taking care of the order of \( \hat{a}_i \) and \( \hat{a}_i^\dagger \), we obtain the Hamiltonian operator of the electromagnetic field as

\[ \hat{H} = \sum_{\ell} \hbar \Omega_\ell \left( \hat{n}_\ell + \frac{1}{2} \right), \]  

(32.31)

where \( \hat{n}_\ell \equiv \hat{a}_\ell^\dagger \hat{a}_\ell \) denotes the number operator. The contribution 1/2 arises from the commutation relations and results in the familiar zero-point energy. Since there are infinitely many modes, the zero-point energy becomes infinite, but in general we drop this contribution by shifting the vacuum energy, which does not influence the dynamics. The quantization procedure is completed by writing the vector potential in terms of \( \hat{a}_\ell \) and \( \hat{a}_\ell^\dagger \) as a field operator

\[ \hat{A}(\vec{r}, t) = \sum_{\ell} \frac{2\pi\hbar c^2}{\sqrt{\Omega_\ell}} (\hat{a}_\ell \hat{u}_\ell + \hat{a}_\ell^\dagger \hat{u}_{\ell^*}). \]  

(32.32)

Thus, \( \hat{a}_\ell^\dagger \) is called the creation operator for a photon specified in \( \ell \) as corresponding to the quantum-mechanical excitations of the electromagnetic field, while \( \hat{a}_\ell \) is interpreted as the annihilation operator for a photon in state \( \ell \). It is important to note that the \( \vec{r} \) and \( t \) that appear in the quantized field \( \hat{A}(\vec{r}, t) \) are not quantum-mechanical variables but just parameters on which the field operator depends, and, in particular, \( \vec{r} \) and \( t \) should not be regarded as the space–time coordinates of the photon.

When we adopt a linearly polarized plane wave as the mode function

\[ \hat{u}_\ell(\vec{r}) = \vec{e}_{\ell\alpha} e^{i \vec{\ell} \cdot \vec{r}} \]  

(32.33)

with the polarization vector \( \vec{e}_{\ell\alpha} \), the energy and momentum of the photon are \( \hbar \Omega_\ell = \hbar |\vec{\ell}| \) and \( \hbar \vec{\ell} \), respectively. Therefore, the mass of the photon is zero. In addition, since \( \vec{e}_{\ell\alpha} \) transforms like a vector, the general theory of angular momentum encourages us to associate with it one unit of angular momentum, which means that the photon has one unit of spin angular momentum.

The field operators described above operate on quantum state vectors, and quantum states \( |\Psi\rangle \) of the electromagnetic field are, in general, multimode states that involve quantum states \( |\psi_\ell\rangle \) for each mode \( \ell \). One of the useful quantum states is photon number states denoted by \( |n_\ell\rangle \), which are eigenstates of the number operator \( \hat{n}_\ell \)

\[ \hat{n}_\ell |n_\ell\rangle = n_\ell |n_\ell\rangle \]  

(32.34)

with integer eigenvalues \( n_\ell \). At the same time, \( |n_\ell\rangle \) are eigenstates of the Hamiltonian with eigenenergy \( n_\ell \hbar \Omega_\ell \), that is, \( n_\ell \) times the fundamental unit \( \hbar \Omega_\ell \). It should be noted that \( n_\ell \) quanta of energy \( \hbar \Omega_\ell \) are in the mode, but the energy is distributed over the entire space, that is, not localized. Other useful quantum states used later are coherent states \( |\alpha\rangle \), which are eigenstates of the annihilation operator \( \hat{a}_\ell \) with eigenvalues \( \alpha \)

\[ \hat{a}_\ell |\alpha\rangle = \alpha |\alpha\rangle. \]  

(32.35)

The phase of the coherent states is completely determined, while the number of photons is completely undetermined.

In the subsequent sections, we will employ a quantum electromagnetic field discussed above in order to discuss the nature of light–matter interactions apparently exhibited at the nanoscale.

32.3 Dressing Mechanism and Spatial Localization of Photons

32.3.1 Virtual Photon Cloud Surrounding a Neutral Source (in Ground State or Excited State) in QED

A quantum source material system interacting with a quantum field is influenced by virtual processes such as emission and absorption of virtual quanta of the field, and the source can be described as a dressed source, that is, the “bare” source surrounded by a cloud of virtual particles (Compagno et al. 1988, 1995). It is true for a detector. The virtual-cloud effects are responsible for the modification of the values of fundamental constants. For example, in nonrelativistic quantum electrodynamics, the presence of a virtual cloud around a hydrogen atom in its ground state contributes to the Lamb shift. Dressed-source effects can also be seen in different physical systems, such as a nucleon coupled to the meson field, and an electron coupled to the optical phonon modes of a semiconductor (polaron). The virtual cloud around the source also modifies the energy density distribution of the electromagnetic field, and the detailed properties have direct physical significance. The energy density of the virtual photon cloud at a given point is in fact related to the van der Waals interaction experienced by a suitable test
object as a detector at that point. The presence of a virtual cloud around a source can influence not only its energy levels but also its dynamics.

Let us roughly estimate the linear dimensions of the virtual cloud surrounding the source or the detector. Even when the source–field system is in the ground state, a fluctuation of the field leads to the possibility of absorption or the emission of photons, not necessarily to the conservation of energy. Such energy imbalance $\Delta E$ is constrained by the Heisenberg uncertainty principle $\Delta E \sim h/\tau$, where $\tau$ is the duration of the fluctuation. Since these fluctuations take place continuously, a steady-state cloud of virtual photons is continuously emitted and reabsorbed. The virtual photon can only attain a finite distance from the source or the detector given by

$$r \sim c \tau \sim \frac{hc}{\Delta E},$$  \hspace{1cm} (32.36)

where $c$ is the speed of light. For a transition corresponding to one of typical visible light, we set $\Delta E \sim 2eV$ and obtain a typical linear dimension of 100 nm. This indicates that dressing effects might be prominent at the nanoscale.

We have discussed the virtual clouds of the source or detector in its ground state. From now on, we discuss the virtual cloud of the source or the detector in excited states, which can decay by emission of real photons. The above discussion inclines us to use the perturbation theory, but it fails due to the denominators at all the orders of perturbation theory. Another attempt way. One of such attempts is based on an extension of dressing effects, not necessarily to the conservation of energy. Such energy imbalance $\Delta E$ is constrained by the Heisenberg uncertainty principle $\Delta E \sim h/\tau$, where $\tau$ is the duration of the fluctuation. Since these fluctuations take place continuously, a steady-state cloud of virtual photons is continuously emitted and reabsorbed.

In these kinds of studies, the multipolar quantum dynamics in Coulomb gauge is employed because all the interactions, except for the Coulomb binding within each molecule, are mediated by transverse photons, and at the same time the retarded effects are automatically satisfied (Power and Thirunamachandran 1993). For example, for upward transitions from the ground state $|0\rangle$ to an excited state $|m\rangle$, the electric–electric spatial correlation expectation value after spatial averaging of the molecular orientation is given by using the second-order perturbation method to include virtual-cloud effects:

$$\langle D_i(\vec{r}) D_j(\vec{r}) \rangle_{m=0} \sim \begin{cases} \frac{\vec{\mu}_m}{3\pi} (\delta_{ij} + \vec{r}_i \cdot \vec{r}_j), & \text{for near zone (}k_0r \ll 1\text{)}, \\ \frac{\vec{\mu}_m}{6\pi k_0^2} (13\delta_{ij} + 7\vec{r}_i \cdot \vec{r}_j), & \text{for far zone (}k_0r \gg 1\text{)} \end{cases}$$  \hspace{1cm} (32.37)

where $D_i(\vec{r})$ is the $i$-component of the transverse displacement vector field $\vec{D}(\vec{r})$, which satisfies $\nabla \cdot \vec{D}(\vec{r}) = 0$, $\vec{\mu}_m$ is the electric dipole transition moment for the molecular states $|m\rangle$ and $|0\rangle$, and $k_0 \equiv k_{mn} = (E_m - E_0)/(hc)$ denotes the wave number associated with the $m \leftarrow 0$ transition of the molecule. The $i$-component of the unit vector $\vec{r}$ is designated by $\vec{r}_i$, while the absolute value of the position vector $\vec{r}$ is expressed by $r$. The Kronecker delta is denoted by $\delta_{ij}$.

Similarly, for downward transitions from an excited state $|p\rangle$ to the ground state $|0\rangle$, the electric–electric correlation function is obtained:

$$\langle D_i(\vec{r}) D_j(\vec{r}) \rangle_{i=p} \sim \begin{cases} \frac{2p_0^2 \vec{\mu}_p}{3\pi^2} (\delta_{ij} - \vec{r}_i \cdot \vec{r}_j), & \text{for far zone (}p_0r \gg 1\text{)}, \\ \frac{\vec{\mu}_p}{3\pi^2} (\delta_{ij} + \vec{r}_i \cdot \vec{r}_j), & \text{for near zone (}p_0r \ll 1\text{)} \end{cases}$$  \hspace{1cm} (32.38)

where $p_0 \equiv p_{mp} = (E_p - E_0)/(hc)$ denotes the wave number associated with the $0 \leftarrow p$ transition of the molecule. It should be noted that the $r^{-2}$ dependence arises from the real photon emission, while the $r^{-6}$ dependence and the $r^{-7}$ dependence are due to the virtual photon exchange. The far-zone behavior for the magnetic–magnetic correlation functions due to an electric-dipole source is also described by the same $r^{-2}$ or $r^{-7}$ dependence, while the near-zone result is different from its electric analog and the power law is $r^{-3}$ instead of $r^{-6}$.

We have discussed the electric and magnetic correlation functions leading to the electric and magnetic energy densities associated with electric-dipole transitions in a source molecule, which can be detected by their effect on polarizable test bodies placed in the neighborhood of the source. This situation is analogous to an optical near-field system in which a nanometric material source connected to a macroscopic material and light source...
interacts with a nanometric detector connected to a macroscopic detector system. The difference is that the nanometric detector serving as a test body can disturb the field formed by the nanometric source in the case of the optical near-field system. We will move on to this topic in Section 32.3.3.

32.3.3 Effective Near-Field Optical Interaction between Nanomaterials Disconnected but Closely Separated

Several theoretical approaches to optical near-field problems, different from each other in viewpoints, have been proposed in the last two decades (Pohl and Courjon 1993, Ohtsu and Hori 1999). The optical near-field problems, including the application to nanophotonics, are ultimately regarded as how one should formulate a separated (more than two elements) composite system, each of which consists of a photon–electron–phonon interacting system on a nanometer scale as a source or a detector system and, which, at the same time, is connected with a macroscopic light–matter system. These questions must be clearly answered to achieve practical realization of nanophotonics. In order to provide a base for a variety of discussions in this research field, a new formulation has been developed within a quantum theoretical framework, putting matter excitations (electronic and vibrational) on an equal footing with photons (Kobayashi et al. 2001).

As discussed in Section 32.2, a “photon,” whose concept has been established as a result of quantization of a free electromagnetic field, corresponds to a discrete excitation of electromagnetic modes in a virtual cavity. Unlike an electron, a photon is massless, and it is difficult to construct a wave function in the position representation that gives a picture of the photon as a spatially localized point particle like an electron. However, if there is a detector near the optical source, such as an atom, to absorb a photon in an area whose linear dimension is much smaller than the wavelength of light, it would be possible to detect a photon with the same precision as the detector size. In optical near-field problems, it is required to consider the interactions between light and nanomaterials surrounded by a macroscopic material system and detection of light by other nanomaterials on a nanometer scale. Then, a more serious question for the quantization of the field is how to define a virtual cavity, or which normal modes are to be used, since there exist more than two systems (nano-source and nano-detector)—either one is in the excited state), as a result of renormalizing the other effects. It corresponds to an approach to describe “photons localized around nanomaterials,” as if each nanomaterial would work as a detector and light source in a self-consistent way. The effective interaction related to optical near-fields is hereafter called a near-field optical interaction. As will be discussed in detail in this section, the near-field optical interaction between nanomaterials separated by $R$ is as follows:

$$V_{\text{eff}} = \frac{\exp(-aR)}{R},$$

(32.39)

where $a^{-1}$ is the interaction range that represents the characteristic size of the nanomaterials and does not depend on the wavelength of the light used. It indicates that photons are localized around the nanomaterials (either of which is in the excited state) as a result of the interaction with matter fields, from which a photon, in turn, can acquire a finite mass. Therefore, we might consider that the near-field optical interaction is produced via localized photon hopping between nanomaterials. On the basis of the projection operator method, we will investigate formulation of an optical near-field system that was briefly mentioned above. Moreover, the explicit functional form of the near-field optical interaction will be given by using the effective interaction $m\tilde{V}_{\text{eff}}$ in a perturbative way.

32.3.3.1 Relevant Nanometric Subsystem and Irrelevant Macroscopic Subsystem

As schematically illustrated in Figure 32.1, the optical near-field system consists of two subsystems: One is a macroscopic subsystem including the incident light, whose typical dimension is much larger than the wavelength of the incident light. The other is a nanometric subsystem (nano-source and nano-detector), whose constituents are, for example, a nanometric aperture or a protrusion at the apex of a near-field optical probe, and a nanometric sample. We call such an aperture or a protrusion a probe tip.
As a nanometric sample we mainly suppose a single atom/molecule, or quantum dot (QD). Two subsystems are interacting with each other, and it is very important to formulate the interaction consistently and systematically. Here let us call the nanometric subsystem a relevant subsystem \( n \), and the macroscopic subsystem an irrelevant subsystem \( M \). We are interested in the subsystem \( n \); in particular, the interaction induced in the subsystem \( n \). Therefore, it is the key to renormalize the effects originating from the subsystem \( M \) in a consistent and systematic way. Now we show a formulation based on the projection operator method, described below.

### 32.3.3.2 P Space and Q Space

It is preferable, for a variety of discussions, to express exact eigenstates \( |\psi\rangle \) for the total system described by the total Hamiltonian \( \hat{H} \) in terms of a small number of bases of as small a number of degrees of freedom as possible, which span P space. In the following, let us assume two states as the P-space components: \(|\phi\rangle = |s\rangle |p\rangle \otimes |0_{(M)}\rangle \) and \(|\phi\rangle = |s\rangle |p\rangle \otimes |0_{(M)}\rangle \), both of which are eigenstates of the unperturbed Hamiltonian \( \hat{H}_0 \). Here \(|s\rangle \) and \(|p\rangle \) are eigenstates of the sample that is isolated from the others, while \(|p\rangle \) and \(|p\rangle \) are eigenstates of the probe tip, which is also isolated. In addition, exciton polariton states, which are a mixture of photons and electron–hole pairs, are used as bases to describe the macroscopic subsystem \( M \) and thus \(|0_{(M)}\rangle \) represents the vacuum for exciton polaritons. Note that there exist photons and electronic matter excitations even in the vacuum state \(|0_{(M)}\rangle \). The direct product is denoted by the symbol \( \otimes \). The complementary space to the P space is called Q space, which is spanned by a huge number of bases of a large number of degrees of freedom not included in the P space, as schematically shown in Figure 32.2.

### 32.3.3.3 Effective Interaction Exerted in the Nanometric Subsystem

Noticing the relation between a bare interaction Hamiltonian \( \hat{V} = \hat{H} - \hat{H}_0 \) and an effective interaction Hamiltonian operator \( \hat{V}_{\text{eff}} \), namely, \( \langle \psi | \hat{V} | \psi \rangle = \langle \psi | \hat{V}_{\text{eff}} | \psi \rangle \), we obtain the effective interaction Hamiltonian operator in the P space, given by

\[
\hat{V}_{\text{eff}} = (Pj^j \hat{P})^{-1/2} (Pj^j \hat{V} \hat{P})(Pj^j \hat{P})^{-1/2},
\]

and tracing out the other degrees of freedom gives an effective interaction Hamiltonian of the nanometric subsystem \( n \) after renormalizing the effects from the macroscopic subsystem \( M \). Here, \( \hat{P} \) is the projection operator, and \( \hat{Q} \) is the complimentary operator defined by \( \hat{Q} = 1 - \hat{P} \), both of which satisfy the following relations:

\[
P = P^\dagger, \quad P^2 = P, \quad [P, \hat{H}_0] = 0,
\]

\[
Q = Q^\dagger, \quad Q^2 = Q, \quad [Q, \hat{H}_0] = 0,
\]

\[
PQ = QP = 0.
\]

The operator \( \hat{j} \) is defined by

\[
\hat{j} = [1 - (E - \hat{H}_0)^{-1} Q \hat{V}]^{-1},
\]

where \( E \) are the eigenvalues of the total Hamiltonian \( \hat{H} \). Using the effective interaction Hamiltonian, one can forget the subsystem \( M \) as if the subsystem \( n \) were isolated and separated from the subsystem \( M \).

To obtain an explicit expression of the effective interaction Hamiltonian, let us employ the bare interaction \( \hat{V} \) between the two subsystems, which in the multipolar formalism (Craig and Thirunamachandran 1998) is given by

\[
\hat{V} = -\left\{ \hat{\mu}_s \cdot \hat{D}^\dagger (\vec{r}) + \hat{\mu}_p \cdot \hat{D}^\dagger (\vec{r}) \right\},
\]

where the canonical momentum of the vector potential operator \( \hat{A} (\vec{r}) \) is proportional to the transverse displacement vector field operator \( \hat{D}^\dagger (\vec{r}) \), while the electric dipole operator is denoted as \( \hat{\mu} (\vec{r}) \). It should be noted that there are no interactions, i.e., \( \hat{V} = 0 \), without incident photons in the macroscopic subsystem \( M \). The subscripts \( s \) and \( p \) represent physical quantities related to the sample and the probe tip, respectively. Representative positions of the sample and the probe tip are chosen, for simplicity, by the vectors \( \vec{r}_s \) and \( \vec{r}_p \), respectively, but may be composed of several positions. In that case the quantities inside curly brackets in (32.43) should be read as a summation. The operator \( \hat{\Pi} (\vec{r}) \) conjugate to \( \hat{A} (\vec{r}) \) is expressed in terms of \( \hat{D}^\dagger (\vec{r}) \) as follows:

* The transverse component is defined by \( \nabla \cdot \hat{D}^\dagger = 0 \), while the longitudinal component is defined by \( \nabla \times \hat{D}^\dagger = 0 \), for an arbitrary vector field \( \vec{F} (\vec{r}) \).
\[ \hat{\Pi}(\vec{r}) = -\frac{1}{4\pi c} \hat{E}^z(\vec{r}) - \frac{1}{c} \hat{\tilde{P}}^z(\vec{r}) = -\frac{1}{4\pi c} \hat{D}^z(\vec{r}), \]

where \( \hat{E}^z(\vec{r}) \) and \( \hat{\tilde{P}}^z(\vec{r}) \) are the transverse electric field and the induced polarization field, respectively. With the help of the mode expansion of \( \hat{\Lambda}(\vec{r}) \) and \( \hat{\Pi}(\vec{r}) \), that is,

\[ \hat{\Lambda}(\vec{r}) = \sum_{k} \sum_{\lambda=1}^{2} \left( \frac{2\pi\hbar c}{V\omega_k} \right)^{1/2} \hat{e}_\lambda(k) \left\{ \hat{a}_\lambda^\dagger(k)e^{i\vec{k}\cdot\vec{r}} + \hat{a}_\lambda(k)e^{-i\vec{k}\cdot\vec{r}} \right\}, \]

(32.45)

and

\[ \hat{\Pi}(\vec{r}) = -\frac{i}{4\pi c} \sum_{k} \sum_{\lambda=1}^{2} \left( \frac{2\pi\hbar\omega_k}{V} \right)^{1/2} \hat{e}_\lambda(k) \left\{ \hat{a}_\lambda(k)e^{i\vec{k}\cdot\vec{r}} - \hat{a}_\lambda^\dagger(k)e^{-i\vec{k}\cdot\vec{r}} \right\}, \]

(32.46)

we can rewrite the transverse component of the electric displacement operator as

\[ \hat{\tilde{D}}^z(\vec{r}) = i \sum_{k} \sum_{\lambda=1}^{2} \left( \frac{2\pi\hbar\omega_k}{V} \right)^{1/2} \hat{e}_\lambda(k) \left\{ \hat{a}_\lambda(k)e^{i\vec{k}\cdot\vec{r}} - \hat{a}_\lambda^\dagger(k)e^{-i\vec{k}\cdot\vec{r}} \right\}, \]

(32.47)

where the plane waves are used for the mode functions, and the creation and annihilation operators of a photon with wave vector \( \vec{k} \), angular frequency \( \omega_k \), and polarization component \( \lambda \) are designated by \( \hat{a}_\lambda(k) \) and \( \hat{a}_\lambda^\dagger(k) \), respectively. The quantization volume is \( V \), and the unit vector related to the polarization direction is shown by \( \hat{e}_\lambda(k) \). Note that the electric field outside the material corresponds to \( \hat{\tilde{D}}^z(\vec{r}) \).

Since exciton polariton states are employed as bases to describe the macroscopic subsystem \( M \), the creation and annihilation operators of a photon in (32.47) are rewritten using the exciton polariton operators \( \hat{\xi}_\lambda(\vec{k}) \) and \( \hat{\tilde{\xi}}_\lambda(\vec{k}) \), and then they are substituted into (32.43). Using the electric dipole operator defined by

\[ \hat{\tilde{\mu}}_\alpha = \left( \hat{B}(\vec{r}) + \hat{B}^\dagger(\vec{r}) \right) \hat{\mu}_\alpha, \]

(32.48)

with the creation and annihilation operators of excitation in subsystem \( n \), \( \hat{B}(\vec{r}_n) \) and \( \hat{B}^\dagger(\vec{r}_n) \), and the transition dipole moments \( \hat{\mu}_\alpha(\alpha = s, p) \), we obtain the bare interaction in the exciton polariton picture:

\[ \hat{V} = -\sum_{\alpha=s,p} \sum_{\vec{k}} \left( \frac{2\pi\hbar}{V} \right)^{1/2} \left\{ \hat{B}(\vec{r}_n) + \hat{B}^\dagger(\vec{r}_n) \right\} \left( K_{\alpha}(\vec{k}) \hat{\xi}_\lambda(\vec{k}) - K_{\alpha}^\dagger(\vec{k}) \hat{\xi}^\dagger_\lambda(\vec{k}) \right). \]

(32.49)

Here \( K_{\alpha}(\vec{k}) \) is the coupling coefficient between the exciton polariton and the nanometric subsystem \( n \), given by

\[ K_{\alpha}(\vec{k}) = \sum_{i=1}^{2} \left( \hat{\xi}_\alpha^i(\vec{k}) \right) f(k)e^{i\vec{k}\cdot\vec{r}_n}. \]

(32.50)

with

\[ f(k) = \frac{ck}{\sqrt{\Omega^2(k) - \omega^2 - (ck)^2}}. \]

(32.51)

The complex conjugate of \( K_{\alpha}(\vec{k}) \) is denoted by \( K_{\alpha}^*(\vec{k}) \), while \( c, \Omega(k) \), and \( \Omega \) are light speed in vacuum and the eigenfrequencies of both exciton polariton and electronic polarization of the macroscopic subsystem \( M \), respectively. The dispersion relation for a free photon, \( \omega = \Omega(k) \), is used in (32.51). Note that the wave-number dependence of \( f(k) \) characterizes a typical interaction range of exciton polaritons coupled to the nanometric subsystem \( n \).

The next step is to evaluate the amplitude of the effective interaction exerted on the nanometric subsystem, for example, the effective sample–probe interaction in the P space after tracing out the polariton degrees of freedom:

\[ V_{\text{eff}}(2,1) = \langle \phi_i | \hat{V}_{\text{eff}} | \phi_1 \rangle. \]

(32.52)

With the first-order approximation \( \hat{J}^{(1)} \) in (32.42), we can explicitly write (32.52) in the following form:

\[ V_{\text{eff}}(2,1) = \langle \phi_i | \hat{P} \hat{V} \hat{Q} \hat{V} | \phi_1 \rangle. \]

(32.53)

The second line shows that a virtual transition from the initial state \( | \phi_i \rangle \) in the P space to an intermediate state \( | m \rangle \) in the Q space is followed by a subsequent virtual transition from the intermediate state \( | m \rangle \) to the final state \( | \phi_1 \rangle \) in the P space. Here \( E^\text{P}_1(E^\text{P}_2) \) and \( E^\text{Q}_m \) denote eigenenergies of \( | \phi_i \rangle | \phi_1 \rangle \) in the P space and that of \( | \phi_m \rangle \) in the Q space, respectively. Now we can proceed to the next step by substituting the explicit bare interaction \( \hat{V} \) in (32.49) with (32.50) and (32.51) into (32.53). First of all, note that the one-exciton polariton state among arbitrary intermediate states \( | m \rangle \) can only contribute to nonzero matrix elements. Therefore, (32.53) can be transformed into

\[ V_{\text{eff}}(2,1) = -\frac{\hbar}{(2\pi)^2} \int d^d\vec{k} \left[ \frac{K_{\alpha}(\vec{k})K_{\alpha}^*(\vec{k})}{\Omega(\vec{k}) - \Omega(s)} + \frac{K_{\alpha}(\vec{k})K_{\alpha}^*(\vec{k})}{\Omega(\vec{k}) + \Omega(s)} \right]. \]

(32.54)

where the summation over \( \vec{k} \) is replaced by \( \vec{k} \)-integration, i.e.,

\[ \frac{V}{(2\pi)^3} \int d^d\vec{k}, \]

in the usual manner. Excitation energies of the
sample (between $|s\rangle$ and $|\phi\rangle$) and the probe tip (between $|p\rangle$ and $|\psi\rangle$) are denoted as $E_s = h\Omega_s(s)$ and $E_p = h\Omega_p(\rho)$, respectively.

Exchanging the arguments 1 and 2, or the role of the sample and probe tip, we can similarly calculate $V_{\text{eff}}(1,2) = \langle \phi|V|\psi\rangle$:

$$V_{\text{eff}}(1,2) = -\frac{\hbar}{2\pi} \int d^3k \left[ \frac{K_s(k)K_p^*(k)}{\Omega(k) - \Omega_s(p)} + \frac{K_p(k)K_s^*(k)}{\Omega(k) + \Omega_p(\rho)} \right].$$

(32.55)

Therefore, the total amplitude of the effective sample–probe-tip interaction is given by the sum of (32.54) and (32.55), which includes the effects from the macroscopic subsystem $M$. We write this effective interaction for the nanoscopic subsystem $n$ as $V_{\text{eff}}(\hat{r})$ in the following way:

$$V_{\text{eff}}(\hat{r}) = -\frac{1}{4\pi^2} \sum_{\ell=1}^2 \sum_{\alpha=p,\rho} \int d^3k \left[ (\hat{m}_\ell \cdot \hat{c}_{\ell,\lambda}(\hat{k})) (\hat{m}_\ell \cdot \hat{c}_{\lambda,\lambda}(\hat{k})) \right] \times \hbar \omega(\hat{k}) \left( \frac{e^{i\delta_{\alpha}}}{E(k) + E_s} + \frac{e^{-i\delta_{\alpha}}}{E(k) - E_s} \right)$$

(32.56)

where we have set $E(k) = h\Omega(k)$, and $E_\alpha = h\Omega_\alpha(\alpha) - h\Omega_\alpha(\alpha)$ for $\alpha = p$ and $\alpha = s$. The summation over polarization $\lambda$ is performed as

$$\sum_{\lambda=1}^2 \hat{c}_{\lambda,\lambda}(\hat{k})\hat{c}_{\lambda}(\hat{k}) = \delta_\alpha - k\hat{k}_\lambda,$$

(32.57)

and thus the summation of $(\hat{m}_\ell \cdot \hat{c}_{\ell,\lambda}(\hat{k})) (\hat{m}_\ell \cdot \hat{c}_{\lambda,\lambda}(\hat{k}))$ over $\lambda$ can be reduced as follows:

$$\sum_{\lambda=1}^2 (\hat{m}_\ell \cdot \hat{c}_{\ell,\lambda}(\hat{k})) (\hat{m}_\ell \cdot \hat{c}_{\lambda,\lambda}(\hat{k})) = \sum_{\lambda=1}^2 \sum_{i,j} (\hat{m}_\ell \hat{c}_{\ell,\lambda}(\hat{k})) (\hat{m}_\ell \hat{c}_{\lambda,\lambda}(\hat{k}))$$

$$= \sum_{i,j} (\hat{m}_\rho \hat{c}_{\rho,\lambda}(\hat{k})) (\hat{m}_\rho \hat{c}_{\lambda,\lambda}(\hat{k}))$$

(32.58)

with the unit vector $\hat{k} = \hat{k}$. Noticing that $d^3k = k^2dkd\Omega = k^2dk \sin\theta d\theta d\phi$ and

$$\delta_\alpha \int_0^1 e^{i\delta_{\alpha}} d\Omega = \delta_\alpha \int_0^1 e^{i\delta_{\alpha}} \cos\theta d\Omega \sin\theta d\phi = \delta_\alpha \frac{2\pi}{ikr} (e^{i\delta_{\alpha}} - e^{-i\delta_{\alpha}}),$$

(32.59a)

we find

$$\int \delta_\alpha \hat{k} \hat{k} e^{i\delta_{\alpha}} d\Omega = \frac{1}{k^2} \nabla \nabla \cdot e^{i\delta_{\alpha}} d\Omega = \frac{2\pi}{ik^3} \nabla \nabla \cdot \left( \frac{e^{i\delta_{\alpha}} - e^{-i\delta_{\alpha}}}{r} \right).$$

(32.59b)
32.3.4 Localization of a Photon Dressed by Matter Excitation in Nanomaterials at the Nanoscale

In this section, we consider a simple model system, for example, a pseudo one-dimensional near-field optical probe system, to discuss the mechanism of photon localization in space as well as the phonon’s role. In order to focus on the photon–phonon interaction, the interacting part between photon and electronic excitation is first expressed in terms of a polariton, and is called a photon in the model. Then the model Hamiltonian that describes the photon and phonon interacting system is presented. Using the Davydov transformation, we rewrite the Hamiltonian in terms of quasiparticles. On the basis of the Hamiltonian, we present numerical results on the spatial distribution of photons and discuss the mechanism of photon localization due to phonons.

32.3.4.1 Model Hamiltonian

We consider a near-field optical probe, schematically shown in Figure 32.3, as an example system where light interacts with both phonons and electrons in the probe on a nanometer scale. Here, the interaction of a photon and an electronic excitation is assumed to be expressed in terms of a polariton basis, as discussed above, and is hereafter called a photon so that special attention is paid to the photon–phonon interaction. The system is simply modeled as a one-dimensional atomic or molecular chain coupled with photon and phonon fields. The chain consists of finite N molecules (representatively called), each of which is located at a discrete point (called a molecular site) whose separation represents a characteristic scale of the near-field system. Photons are expressed in the site representation and can hop to the nearest neighbor sites due to the short-range interaction nature of the optical near fields.

The Hamiltonian for the above model is given by

\[ H = \sum_{i=1}^{N} \hbar \omega_0 \hat{a}_i^\dagger \hat{a}_i + \sum_{i=1}^{N} \frac{\hbar}{2m_e} \left( \hat{x}_{i+1} - \hat{x}_i \right)^2 + \sum_{i=1}^{N} \frac{k}{2} \hat{x}_i^2 + \sum_{i=1}^{N} \hbar f \left( \hat{a}_i^\dagger \hat{a}_{i+1} + \hat{a}_i \hat{a}_{i+1}^\dagger \right), \]  

where

\[ \hat{a}_i^\dagger \] and \( \hat{a}_i \) correspondingly denote the creation and annihilation operators of a photon with energy \( \hbar \omega_0 \) at site \( i \) in the chain.

\( \hat{x}_i \) and \( \hat{p}_i \) represent the displacement and conjugate momentum operators of the vibration, respectively.

**FIGURE 32.3** Simple one-dimensional model for a light–matter interacting system on a nanometer scale.
The mass of a molecule at site $i$ is designated by $m_i$, and each molecule is assumed to be connected by springs with spring constant $k$. The third and fourth terms in (32.66) stand for the photon–vibration interaction with coupling constant $\chi$ and the photon hopping with hopping constant $J$, respectively. After the vibration field is quantized in terms of phonon operators of mode $p$ and frequency $\Omega_p$, $\hat{b}_p^\dagger$ and $\hat{b}_p$, the Hamiltonian (32.66) can be rewritten as

$$
\hat{H} = \sum_{i=1}^{N} \hbar \omega_i \hat{a}_i^\dagger \hat{a}_i + \sum_{p=1}^{N} \hbar \Omega_p \hat{b}_p^\dagger \hat{b}_p + \sum_{i=1}^{N} \sum_{p=1}^{N} h \chi_p \delta \delta \hat{a}_i^\dagger \hat{a}_i (\hat{b}_p^\dagger + \hat{b}_p) + \sum_{i=1}^{N} \sum_{p=1}^{N} \hbar J (\hat{a}_i^\dagger \hat{a}_{i+1} + \hat{a}_i \hat{a}_{i+1}^\dagger),
$$

with the coupling constant $\chi_p$ of a photon at site $i$ and a phonon of mode $p$. It should be noted that the index $p$ designates not the momentum but the mode number, because the translational invariance of the system is broken and the momentum is not a good quantum number. The site-dependent coupling constant $\chi_p$ is related to the original coupling constant $\chi$ in terms of the transformation matrix as $P_p$ as

$$
\chi_p = \chi P_p \sqrt{\frac{\hbar}{2 m_i \Omega_p}},
$$

and the creation and annihilation operators of a photon and a phonon satisfy the boson commutation relation as $[\hat{a}_i, \hat{a}_i^\dagger] = \delta_{ij}$ and $[\hat{b}_p, \hat{b}_p^\dagger] = \delta_{pq}$. The Hamiltonian (32.67), which describes the model system, is not easily handled because of the third order of the operators in the interaction term. To avoid this difficulty, this direct photon–phonon interaction term in (32.67) is eliminated by the Davydov transformation in the following subsection.

### 32.3.4.2 Davydov Transformation

Before going into the explicit expression, we discuss a unitary transformation $U$ generated by an anti-Hermitian operator $\hat{S}$, defined as

$$
\hat{U} \equiv \exp(\hat{S}), \quad \text{with} \quad \hat{U}^\dagger = -\hat{S}
$$

and

$$
\hat{U}^\dagger = \hat{U}^{-1}.
$$

Assume a Hamiltonian $\hat{H}$ that consists of a diagonalized part $\hat{H}_0$ and a non-diagonal interaction part $\hat{V}$:

$$
\hat{H} = \hat{H}_0 + \hat{V}
$$

Transforming the Hamiltonian in (32.70) as

$$
\hat{H} \equiv \hat{U} \hat{H} \hat{U}^\dagger = \hat{U} \hat{H} \hat{U}^{-1},
$$

we have

$$
\hat{H} = \hat{H}_0 + \hat{V} + [\hat{S}, \hat{H}_0] + \frac{1}{2} \{ \hat{S}, [\hat{S}, \hat{H}_0] \} + \cdots.
$$

If the interaction $\hat{V}$ can be perturbative, and if the operator $\hat{S}$ is chosen so that the second and third terms in (32.72) are canceled out, then

$$
\hat{V} = -[\hat{S}, \hat{H}_0],
$$

the Hamiltonian (32.70) is rewritten as

$$
\hat{H} = \hat{H}_0 - \frac{1}{2} \{ \hat{S}, [\hat{S}, \hat{H}_0] \} + \cdots,
$$

and can be diagonalized within the first order of $\hat{V}$.

Now we apply the above discussion to the model Hamiltonian (32.67),

$$
\hat{H}_0 = \sum_{i=1}^{N} \hbar \omega_i \hat{a}_i^\dagger \hat{a}_i + \sum_{p=1}^{N} \hbar \Omega_p \hat{b}_p^\dagger \hat{b}_p,
$$

$$
\hat{V} = \sum_{i=1}^{N} \sum_{p=1}^{N} h \chi_p \delta \delta \hat{a}_i^\dagger \hat{a}_i (\hat{b}_p^\dagger + \hat{b}_p),
$$

tentatively neglecting the hopping term. Assuming the anti-Hermitian operator $\hat{S}$

$$
\hat{S} = \sum_{i} \sum_{p} f_p \hat{a}_i^\dagger \hat{a}_i (\hat{b}_p^\dagger - \hat{b}_p),
$$

we can determine $f_p$ from (32.73) as follows:

$$
f_p = \frac{\chi_p}{\Omega_p}.
$$

This operator form of $\hat{S}$ leads us to not the perturbative but the exact transformation of the photon and phonon operators:

$$
\hat{a}_i^\dagger = \hat{U}^\dagger \hat{a}_i \hat{U} \exp \left\{ - \sum_{p} \frac{\chi_p}{\Omega_p} \left( \hat{b}_p^\dagger - \hat{b}_p \right) \right\},
$$

(32.77a)
The transformed operators can be regarded as the creation and annihilation operators of quasiparticles—dressed photons and phonons—that satisfy the same boson commutation relations as those of photons and phonons before the transformation, namely, 

\[ \hat{\alpha}_i^{\dagger} \hat{\alpha}_i = \hat{U}^\dagger \hat{a}_i \hat{a}_i \hat{U} = \delta_{ii} \quad \text{and} \quad \hat{\beta}_p^{\dagger} \hat{\beta}_p = \hat{U}^\dagger \hat{b}_p \hat{b}_p \hat{U} = \delta_{pp} \]

Using the quasiparticle operators, we can rewrite the Hamiltonian (32.67) as

\[
\hat{H} = \sum_{i=1}^{N} \hbar \omega \hat{\alpha}_i^{\dagger} \hat{\alpha}_i + \sum_{p=1}^{N} \hbar \Omega_p \hat{\beta}_p^{\dagger} \hat{\beta}_p - \sum_{i=1}^{N} \sum_{p=1}^{N} \sum_{q=1}^{N} \frac{\hbar \kappa \Omega_p}{\Omega_{pq}} \hat{\alpha}_i^{\dagger} \hat{\alpha}_i \hat{\alpha}_j \hat{\alpha}_j + \sum_{j=1}^{N} \left( \hat{J}_i \hat{\alpha}_i^{\dagger} \hat{\alpha}_i + \hat{J}_i^{\dagger} \hat{\alpha}_i \hat{\alpha}_i \right),
\]

where it is noted that the direct photon–phonon coupling term has been eliminated, while the quadratic form \( \hat{\alpha}_i^{\dagger} \hat{\alpha}_i \hat{\alpha}_i \hat{\alpha}_i \) has emerged as well as the site-dependent hopping operator \( \hat{J}_i \) in (32.78b). The number states of quasiparticles are then excited by the photon–phonon coupling through the dressed photon hopping. Therefore, it is a more appropriate form to discuss the phonon’s effect on photon’s behavior as localization.

### 32.3.4.3 Quasiparticle and Coherent State

In the previous section, we have transformed the original Hamiltonian with the Davydov transformation. In order to grasp the physical meanings of the quasiparticles introduced above, the creation operator \( \hat{\alpha}_i^{\dagger} \) is applied to the vacuum state \( |0\rangle \). Then, it follows from (32.77a) that

\[
\hat{\alpha}_i^{\dagger} |0\rangle = \hat{a}_i^{\dagger} \exp \left\{ - \sum_{p=1}^{N} \frac{\chi_p}{\Omega_p} (\hat{b}_p^{\dagger} - \hat{b}_p) \right\} |0\rangle = \hat{a}_i^{\dagger} \exp \left\{ - \sum_{p=1}^{N} \frac{1}{2} \left( \frac{\chi_p}{\Omega_p} \right)^2 \exp \left\{ - \frac{\chi_p}{\Omega_p} \hat{b}_p \right\} \right\} |0\rangle.
\]

where a photon at site \( i \) is associated with phonons in a coherent state, i.e., a photon is dressed by an infinite number of phonons. This corresponds to the fact that an optical near field is generated from a result of interactions between the photon and matter fields.

When \( \hat{\beta}_p^{\dagger} \) is applied to the vacuum state \( |0\rangle \), we have

\[
\hat{\beta}_p^{\dagger} |0\rangle = \hat{b}_p^{\dagger} |0\rangle.
\]

and it is expressed by only the bare phonon operator (before the transformation) in the same \( p \) mode. Therefore, we mainly focus on the quasiparticle expressed by \( (\hat{\alpha}_i^{\dagger}, \hat{\alpha}_i) \) in the following subsection. Note that it is valid only if the bare photon number (the expectation value of \( \hat{\alpha}_i^{\dagger} \hat{\alpha}_i \)) is not so large that the fluctuation is more important than the bare photon number. In other words, the model we are considering is suitable for discussing the quantum nature of a few photons in an optically excited probe system.

In the coherent state of the number of phonons, as well as energy, is fluctuating. This fluctuation allows incident photons into the probe system to excite phonon fields. When only two photons in the vacuum state at time \( t = 0 \), the excitation probability \( P(t) \) that a photon incident on site \( i \) in the model system excites the phonon mode \( p \) at time \( t \) is given by

\[
P(t) = 1 - \exp \left\{ - \sum_{p=0}^{N} \exp \left[ - \frac{1}{2} \left( \frac{\chi_p}{\Omega_p} \right)^2 \cos(\Omega_p t) - 1 \right] \right\},
\]

where the photon-hopping term is neglected for simplicity. The excitation probability oscillates at a frequency of \( 2\pi/\Omega_p \), and the maximum value at \( t = \pi/\Omega_p \). The frequencies of the localized phonon modes are higher than those of the delocalized ones, and the localized modes at the earlier time are excited by the incident photons.

Figure 32.4 shows the temporal evolution of the excitation probability \( P_{\beta_i}(t) \) calculated from

\[
P_{\beta_i}(t) = \left[ 1 - \exp \left\{ - \sum_{p=0}^{N} \frac{1}{2} \left( \frac{\chi_p}{\Omega_p} \right)^2 \cos(\Omega_p t) - 1 \right] \right] \exp \left\{ \sum_{p_{\beta_i}} \frac{1}{2} \left( \frac{\chi_p}{\Omega_p} \right)^2 \cos(\Omega_p t) - 1 \right\},
\]

where a specific phonon mode \( \beta_i \) is excited, while other modes are in the vacuum state. In Figure 32.4, the solid curve represents the probability that a localized phonon mode is excited as the \( \beta_i \) mode, while the dashed curve illustrates how the lowest phonon mode is excited as the \( \beta_0 \) mode. It follows from the figure that the localized phonon mode is dominantly excited at an earlier time.
32.3.4.4 Localization Mechanism of Dressed Photons

In this section, we discuss how phonons contribute to the spatial distribution of photons in the pseudo one-dimensional system under consideration. When there are no interactions between photons and phonons, the frequency and hopping constant are equal at all sites, and thus the spatial distribution of photons is symmetric. It means that no photon localization occurs at any specific site. However, if there are any photon–phonon interactions, spatial inhomogeneity or localization of phonons affects the spatial distribution of photons. On the basis of the Hamiltonian (32.78a), we analyze the contribution from the diagonal (the third term) and off-diagonal (the fourth term) parts of the Hamiltonian (32.78a) in order to investigate the localization mechanism of photons.

32.3.4.4.1 Contribution from the Diagonal Part

Let us rewrite the third term of the Hamiltonian (32.78a) with the mean field approximation as

\[ \hat{H} = \sum_{i=1}^{N} \hbar \omega_i \hat{\alpha}_i^\dagger \hat{\alpha}_i + \sum_{i=1}^{N-1} \hbar J \left( \hat{\alpha}_i^\dagger \hat{\alpha}_{i+1} + \hat{\alpha}_{i+1}^\dagger \hat{\alpha}_i \right) \]  (32.84)

or in matrix form as

\[
\hat{H} = \hbar \hat{\alpha}^\dagger \begin{pmatrix}
\omega_- \omega_1 & J & \cdots & 0 \\
J & \omega_- \omega_2 & \cdots & \vdots \\
\vdots & \ddots & \ddots & J \\
0 & \cdots & J & \omega_- \omega_N
\end{pmatrix} \hat{\alpha}
\]  (32.85a)

with

\[ \hat{\alpha} = \left( \hat{\alpha}_1^\dagger, \hat{\alpha}_2^\dagger, \ldots, \hat{\alpha}_N^\dagger \right) \]  (32.85b)

where the effect from the phonon fields is involved in the diagonal elements \( \omega_i \). Denoting an orthonormal matrix to diagonalize the Hamiltonian (32.85a) as \( Q \) and the \( r \)th eigenvalue as \( E_r \), we have

\[ \hat{H} = \sum_{r=1}^{N} \hbar E_r \hat{A}_r^\dagger \hat{A}_r \]  (32.86a)

with

\[ \hat{A}_i = \sum_{r=1}^{N} (Q^{-1})_{ir} \hat{\alpha}_r = \sum_{r=1}^{N} Q_{ir} \hat{\alpha}_r \]  (32.86b)

and

\[
\left[ \hat{A}_i, \hat{A}_j^\dagger \right] = \hat{A}_i \hat{A}_j^\dagger - \hat{A}_j^\dagger \hat{A}_i = \delta_{ij},
\]  (32.86c)

Using the above relations (32.86a) through (32.86c), we can write down the time evolution of the photon number operator \( \hat{N}_i \) at site \( i \) as follows:

\[
\hat{N}_i(t) = \exp \left( \frac{i \hat{H} t}{\hbar} \right) \hat{N}_i \exp \left( -\frac{i \hat{H} t}{\hbar} \right) = \sum_{r=1}^{N} \sum_{s=1}^{N} Q_{ir} Q_{sr} \hat{A}_r^\dagger \hat{A}_s \exp \left\{ i(E_r - E_s)t \right\}.
\]  (32.87)

The expectation value of the photon number operator \( \langle \hat{N}_i(t) \rangle \) is then given by

\[
\langle \hat{N}_i(t) \rangle_j = \langle \psi_j | \hat{N}_i(t) | \psi_j \rangle = \sum_{r=1}^{N} \sum_{s=1}^{N} Q_{ir} Q_{sr} Q_{js} \cos \left\{ (E_r - E_s)t \right\}.
\]  (32.88)
in terms of one photon state at site \( j \) defined by

\[
|\psi_j\rangle = \hat{\alpha}_j |0\rangle = \sum_{r=1}^{N} Q_{jr} |\hat{\alpha}_r\rangle |0\rangle \tag{32.89}
\]

Since the photon number operator \( \hat{N}_i \) commutes with the Hamiltonian (32.84), the total photon number is conserved, which means that a polariton, called a photon in this section, conserves the total particle number within the lifetime. Moreover, \( \langle \hat{N}(t) \rangle \) can be regarded as the observation probability of a photon at an arbitrary site \( i \) and time \( t \), initially populated at site \( j \). This function is analytically expressed in terms of the Bessel functions \( J_{\nu} \):

\[
\langle N_i(t) \rangle_j = \left| J_{\nu}(2\sqrt{\hat{J}t}) - (-1)^{\nu} J_{\nu}(2\sqrt{\hat{J}t}) \right|^2, \tag{32.90}
\]

when there are no photon–phonon interactions (\( \omega = 0 \)) and the total site number \( N \) becomes infinite. Here the argument \( f \) is the photon hopping constant, and (32.90) shows that a photon initially populated at site \( j \) delocalizes to the whole system.

Focusing on the localized phonon modes, we take the summation in (32.83b) over the localized modes only, which means that an earlier stage is considered after the incident photon excites the phonon modes, or that the duration of the localized phonon modes that are dominant over the delocalized modes is focused on (see Figure 32.4). This kind of analysis provides us with an interesting insight into the photon–phonon coupling constant and the photon hopping constant, which is necessary for understanding the mechanism of photon localization.

The temporal evolution of the observation probability of a photon at each site is shown in Figure 32.5. Without the photon–phonon coupling (\( \chi = 0 \)), a photon spreads over the whole system as a result of the photon hopping, as shown in Figure 32.5a. Here the photon energy \( \hbar \omega = 1.81 \text{ eV} \) and the hopping constant \( \hbar J = 0.5 \text{ eV} \) are used in the calculation. Impurities are assumed to be doped at sites 3, 7, 11, 15, and 19, while the total number of sites \( N \) is 20, and the mass ratio of the host molecules to the impurities is 5. Figure 32.5b shows a result with \( \chi = 1.4 \times 10^7 \text{ fs}^{-1} \text{ nm}^{-1} \); the other parameters used are the same as those in Figure 32.5a. It follows from the figure that a photon moves from one impurity site to another impurity site instead of delocalizing to the whole system. As the photon–phonon coupling constant becomes much larger than \( \chi = 1.4 \times 10^7 \text{ fs}^{-1} \text{ nm}^{-1} \), a photon cannot move from the initial impurity site to others, but stays there.

The effect due to the photon–phonon coupling constant \( \chi \) is expressed by the diagonal component in the Hamiltonian, while the off-diagonal component involves the photon hopping effect due to the hopping constant \( J \). The above results indicate that a photon’s spatial distribution depends on the competition between the diagonal and off-diagonal components in the Hamiltonian, i.e., \( \chi \) and \( J \), and that a photon can move among impurity sites and localize at those sites when both components are comparable under the condition

\[
\chi \sim N \sqrt{\frac{kf}{\hbar}}, \tag{32.91}
\]

where the localization width seems very narrow.

### 32.3.4.4.2 Contribution from the Off-Diagonal Part

In the previous subsection, we have approximated \( J \) as a constant independent of the sites, in order to examine the photon’s spatial distribution as well as the mechanism of the photon localization. Now let us treat the photon hopping operator \( \hat{J} \) more rigorously, and investigate the site dependence of the off-diagonal contribution, which includes the inhomogeneity of the phonon fields. Noticing that a quasiparticle transformed from a photon

![FIGURE 32.5](See color insert following page 21-4.) Probability that a photon is found at each site as a function of time (a) without the photon–phonon coupling, and (b) with the photon–phonon coupling comparable to the photon hopping constant.
operator by the Davydov transformation is associated with phonons in the coherent state (see (32.77a) and (32.77b)), we take expectation values of $\hat{J}$ in terms of the coherent state of phonons $|\gamma\rangle$ as

$$J_i = \langle \gamma | \hat{J}_i | \gamma \rangle. \quad (32.92)$$

Here, the coherent state $|\gamma\rangle$ is an eigenstate of the annihilation operator $\hat{b}_p$ with eigenvalue $\gamma_p$ and satisfies

$$\hat{b}_p |\gamma\rangle = \gamma_p |\gamma\rangle. \quad (32.93)$$

Since the difference between the creation and annihilation operators of a phonon is invariant under the Davydov transformation, the following relation holds:

$$\hat{\beta}_p^\dagger - \hat{\beta}_p = \hat{b}_p^\dagger - \hat{b}_p. \quad (32.94)$$

Using (32.78b), (32.93), and (32.94), we can rewrite the site-dependent hopping constant $J_i$ in (32.92) as

$$J_i = J \langle \gamma | \exp \left\{ \sum_{p=1}^{N} C_p (\hat{b}_p^\dagger - \hat{b}_p) \right\} |\gamma\rangle$$

$$= J \exp \left\{ \frac{1}{2} \sum_{p=1}^{N} C_p^2 \right\} \langle \gamma | \exp \left\{ \sum_{p'=1}^{N} C_{p'} \hat{b}_{p'}^\dagger \right\} \exp \left\{ \sum_{p'=1}^{N} C_{p''} \hat{b}_{p''} \right\} |\gamma\rangle$$

$$= J \exp \left\{ \frac{1}{2} \sum_{p=1}^{N} C_p^2 \right\}, \quad (32.95a)$$

where $C_p$ is denoted by

$$C_p = \frac{\chi_{pp} - \chi_{ppp}}{\Omega_p}. \quad (32.95b)$$

Figure 32.6 shows the site dependence of $J_i$ in the case of $N = 20$. Impurities are doped at sites 4, 6, 13, and 19. The mass ratio of the host molecules to the impurities is 1–0.2, whereas $\hbar = 0.5$ eV and $\chi = 40.0 \text{fs}^{-1} \text{nm}^{-1}$ are used. It follows from the figure that the hopping constants are highly modified around the impurity sites and the edge sites. The result implies that photons are strongly affected by localized phonons and hop to the impurity sites to localize. Here we have not considered the temperature dependence of $J_i$, which is important for phenomena dominated by incoherent phonons. This is because coherent phonons weakly depend on the temperature of the system. However, there remains room to discuss a more fundamental issue, i.e., whether the probe system is in a thermal equilibrium state or not.

In Figure 32.7, we present a typical result that photons localize around the impurity sites in the system as the photon–phonon coupling constants $\chi$ vary from 0 to 40.0 $\text{fs}^{-1} \text{nm}^{-1}$ or 54.0 $\text{fs}^{-1} \text{nm}^{-1}$, while keeping $\hbar = 0.5$ eV. As depicted by the filled squares in the figure, photons delocalize and spread over the system without the photon–phonon couplings. When the photon–phonon couplings are comparable to the hopping constants, $\chi = 40.0 \text{fs}^{-1} \text{nm}^{-1}$, photons can localize around the impurity site with a finite width and two sites at half width and half maximum (HWHM), as shown by the filled circles. This finite width of photon localization comes from the site-dependent hopping constants. As the photon–phonon couplings are larger than $\chi = 40.0 \text{fs}^{-1} \text{nm}^{-1}$, photons can localize at the edge sites with a finite width, as well as the impurity sites. In Figure 32.7, the photon localization at the edge site, shown by the filled triangles,
originates from the finite size effect of the molecular chain. This kind of localization of photons dressed by the coherent state of phonons leads us to a simple understanding of phonon-assisted photodissociation using an optical near field: molecules in the electronic ground state approach the probe tip within the localization range of the dressed photons, and can be vibrationally excited by the dressed-photon transfer to the molecules, via a multi-phonon component of the dressed photons, which might be followed by electronic excitation. Thus, it leads to the dissociation of the molecules even if an incident photon energy less than the dissociation energy is used.

As a natural extension of the localized photon model, we have discussed the inclusion of the phonon’s effects into the model. The study was initially motivated by experiments on the photodissociation of molecules by optical near fields, whose results show unique features different from the conventional one with far fields (Ohtsu et al. 2008). After clarifying whether the vibration modes in a pseudo one-dimensional system are delocalized or localized, we focused on the interaction between dressed photons and phonons by using the Davydov transformation. We have theoretically shown that photons are dressed by the coherent state of phonons, and found that the competition between the photon–phonon coupling constant and the photon hopping constant governs the photon localization or delocalization in space. The obtained results lead us to a simple understanding of an optical near field itself as an interacting system of photon, electronic excitation (induced polarization), and phonon fields in a nanometer space, which are surrounded by macroscopic environments, as well as phonon-assisted photodissociation using an optical near field.

### 32.4 Summary and Future Perspective

We have briefly outlined the difficulties in defining the position-representation wave function of a photon, followed by several trials to overcome these issues. On the basis of canonical quantization of the electromagnetic fields in free space, we have discussed a dressing mechanism and spatial localization of photons, which is a natural viewpoint from light–matter interactions, or from virtual photon clouds and field correlations. Finally, with the projection operator method, we have shown an effective interaction between nanomaterials electronically disconnected, but closely separated, which are also surrounded by a macroscopic system, in order to detect the virtual clouds. We have discussed the photon dressing by material excitation and pointed out the importance of the phonon’s role for spatial localization of photons at the nanoscale.

The pace of development in photonics has accelerated, but most of the underlying science of the field is still Maxwell’s classical electromagnetism, not field-quantized photons. In the near future, however, we are anticipating new breakthroughs in nano- and atom photonics, where the localized and quantized nature of photons, as well as an exact quantization formulation for an optical near-field system, including relaxation processes at the nanoscale, will play a critical role.

### Acknowledgments

The author is grateful to M. Ohtsu (University of Tokyo) and H. Hori (University of Yamanashi) for stimulating discussions and valuable comments from the early stage of this study. He is also thankful to S. Sangu (Ricoh Co. Ltd.), A. Shojiguchi (NEC Co.), Y. Tanaka (Tokyo Institute of Technology), and A. Sato (Tokyo Institute of Technology) for discussions and collaborations. He greatly acknowledges the valuable guidance given by M. Tsukada (University of Tokyo, emeritus, Tohoku University), K. Kitahara (Tokyo Institute of Technology, emeritus, International Christian University), M. Kitano (Kyoto University), Y. Masumoto (University of Tsukuba), K. Cho (Osaka University, emeritus), and T. Yabuzaki (Kyoto University, emeritus). Finally but not the least, he expresses his gratitude to H. Ito (Tokyo Inst. Technology). T. Kawazoe (University of Tokyo), T. Yatsui (University of Tokyo), T. Saiki (Keio University), K. Matsuda (Kyoto University), H. Nejo (National Institute of Materials Science), M. Naruse (National Institute of Information and Communication Technology), M. Ikezawa (University of Tsukuba), I. Banno (University of Yamanashi), and H. Ishihara (Osaka Prefecture University).

### References


