

1

Introduction

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1.1

History

Nanophotonics is a novel technology that utilizes the optical near-field, which is the electromagnetic field that mediates the interaction between nanometric particles located in close proximity to each other. The true nature of nanophotonics is to realize ‘qualitative innovation’ in photonic devices, fabrication techniques, and systems by utilizing novel functions and phenomena caused by optical near-field interactions, which are impossible as long as conventional propagating light is used. The author first proposed nanophotonics in 1993 as a way to transcend the diffraction limit, which impedes reducing the size of photonic devices, to improve the resolution of optical fabrication techniques, and increasing the storage density of optical disk memories [1]. Based on his proposal, the Optical Industry Technology Development Association (OITDA) of Japan organized the nanophotonics technical group, and intensive discussions on the future direction of nanophotonics started in April 1994, in collaboration with academia and industry. Although photonic crystals, plasmonics, metamaterials, silicon photonics, and quantum dot lasers have been popular subjects of study in recent years, they are all based on diffraction-limited wave optics. Even if novel or nanometer-sized materials are used for these subjects, the size of a photonic device cannot be reduced beyond the diffraction limit as long as propagating light is used for its operation.

This chapter describes the history and present activities of nanophotonics. Before nanophotonics was founded, the study of optical near-fields started in Japan in the early 1980s, separate from European and American research [2]. Few attended when the author was invited to the first international workshop on near-field optics in 1992 [3], but the numbers have increased very rapidly in subsequent conferences, and near-field optics has become very popular worldwide.

Although the number of near-field optics researchers increased dramatically in the 1990s, almost all of them focused on microscopy or spectroscopy. Unlike them, intensive research and development of novel devices, fabrication techniques, and systems was conducted after nanophotonics was proposed in 1993. To support these studies, a novel theoretical model of optical near-fields was established and applied to the design of nanophotonic devices, fabrication techniques, and systems. For these designs, local energy transfer and its subsequent dissipation are indispensable. They are possible only using optical near-fields [4], which are the elementary surface excitations on nanometric particles, or in the other words, *dressed photons* that carry the material energy. Nanophotonics evolved into related sciences, one of which is atom photonics, which studies the control of the thermal motions of neutral atoms in a vacuum using optical near-fields [5]. The above-mentioned theoretical model has been used as a design criterion for atom photonics for opening technologies in atomic-level material fabrication. Furthermore, atom photonics has triggered related research, such as that seen in the atom chip [6].

1.2

Fiber Probes and Sensing Systems

One decade after near-field optics started in the early 1980s, a reliable, reproducible, and selective chemical etching technology was established for fabricating high-quality fiber probes [7]. This led to the realization of a high-resolution probe with a 1-nm apex radius, a high-efficiency probe with 10% optical near-field generation efficiency, and other devices. Elsewhere in the world, methods that heat and pull the fiber were most popular at that time [8], although the performance of chemically etched fiber probes was superior. The methodology used to fabricate these probes was transferred to industry, and Japanese companies started producing high-quality commercial fiber probes. A near-field optical microscope was developed using these fiber probes, and a variety of ultrahigh-resolution images have been achieved, such as one of a single-stranded DNA molecule with a resolution greater than 4 nm [9], which is a world record.

A near-field spectrometer was also developed for diagnosing single semiconductor quantum dots [10], semiconductor devices [11], a single organic molecule [12], and biological specimens [13]. Many experimental results on spatially resolved photoluminescence and Raman spectra with a 10-nm resolution have been accumulated [14]. Patents have been transferred to industry and a Japanese company has produced commercial near-field photoluminescence spectrometers operating in the ultraviolet–infrared and liquid helium–room temperature ranges [15]. They are popular in a variety of nanoscience and technology fields. Instead of using fiber probes, an apertureless probe was sometimes used because of its simplicity of fabrication and the possibility of field enhancement. However, it was demonstrated that apertureless probes do not realize high resolution due to the scattering of residual propagating light [16].

1.3 Theory

The use of optical near-fields was proposed about 80 years ago as a way to break the diffraction limit [17]. This proposal holds that an optical near-field can be generated on a subwavelength-sized aperture by irradiating the propagating light. It also holds that the size of the spatial distribution of the optical near-field energy depends not on the wavelength of the incident light, but on the aperture size. However, in the early stage of such studies, the concept of optical near-fields was not clearly differentiated from that of an evanescent wave on a planar material surface (i.e. a two-dimensional topographical material) or that of a guided wave in a subwavelength-sized cross-sectional waveguide (i.e. a one-dimensional topographical material). To distinguish these clearly, note that an evanescent wave is generated by primary excitations, that is, electronic dipoles induced near the two-dimensional material surface, which align periodically depending on the spatial phase of the incident light. In contrast, the guided wave in a subwavelength-sized cross-sectional waveguide is generated by the electronic dipoles induced along the one-dimensional waveguide material. They align periodically depending on the spatial phase of the incident light. Silicon waveguides used for silicon photonics and metallic waveguides used for plasmonics are examples. The two-dimensional evanescent wave and one-dimensional guided wave are both diffraction-limited light waves because they are generated by the periodic alignment of electric dipoles depending on the spatial phase of the incident light.

Unlike these waves, an optical near-field is generated by electronic dipoles induced in a nanometric particle (i.e. a subwavelength-sized zero-dimensional topographical material). Their alignment is independent of the spatial phase of the incident light because the particles are much smaller than the wavelength of the incident light. Instead, the optical near-field depends on the size, conformation, and structure of the particles. Due to this independence and dependence, optical science and technology beyond the diffraction limit can be realized only by an optical near-field, and not by an evanescent wave or a guided wave.

Methods such as Green's function, a calculation using the finite-difference time-domain method, etc. have been developed to describe the optical near-field semi-quantitatively based on conventional optics theories [18]. However, conventional optics theories do not provide any physically intuitive pictures of *nonpropagating* nanometric optical near-fields because these theories were developed to describe only the light waves *propagating* through macroscopic space or materials. For nanophotonics, a novel theory has been developed based on a framework that is completely different from those of the conventional theories. This novel theory is based on the interaction and energy transfer between nanometric particles via an optical near-field. This perspective is essential because the interaction, energy transfer, and subsequent dissipation are indispensable for nanophotonic devices and fabrications. That is, to observe a nonpropagating optical near-field, a second particle is inserted to generate observable scattered light by disturbing the optical near-field. However, the real system is more complicated because the *nanometric subsystem* (the two particles and

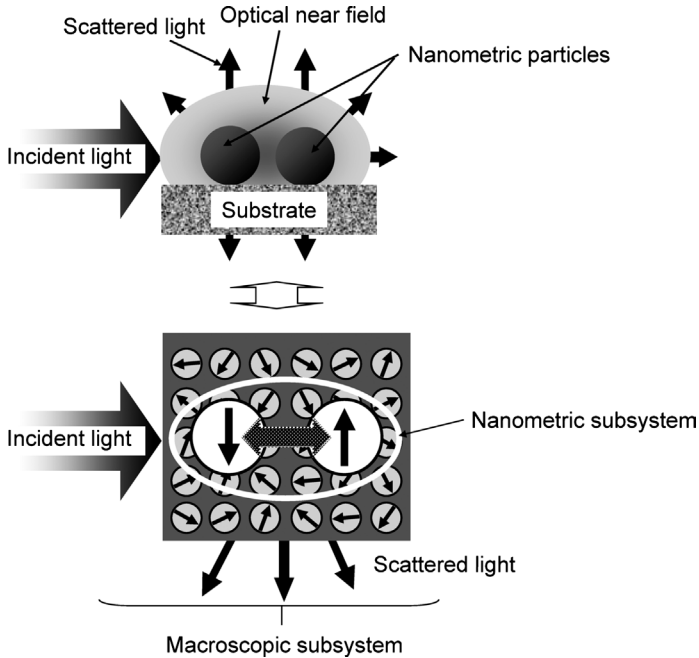


Figure 1.1 A nanometric subsystem composed of two particles and an optical near-field, which are buried in a macroscopic subsystem.

the optical near-field) is buried in a *macroscopic subsystem* consisting of the macroscopic substrate material and the macroscopic electromagnetic fields of the incident and scattered lights (see Figure 1.1).

The premise behind the novel theory is to avoid the complexity in describing all of the behaviors of nanometric and macroscopic subsystems rigorously, since we are interested only in the behavior of the nanometric subsystem. The macroscopic subsystem is expressed as an exciton–polariton, which is a mixed state of material excitation and electromagnetic fields. Since the nanometric subsystem is excited by an electromagnetic interaction with the macroscopic subsystem, the projection operator method is effective for describing the quantum-mechanical states of these systems [4, 19]. Under this treatment, the nanometric subsystem is regarded as being isolated from the macroscopic subsystem, while the functional form and magnitude of effective interactions between the elements of the nanometric subsystem are influenced by the macroscopic subsystem. That is, the two nanometric particles can be considered as being isolated from the surrounding macroscopic system and as interacting by exchanging exciton–polariton energies. Therefore, the optical near-fields can be considered as *dressed photons*, which carry the material energy.

This local electromagnetic interaction takes place so quickly that the uncertainty principle can allow the exchange of a virtual exciton–polariton nonresonantly, as well as the exchange of a real exciton–polariton resonantly (see Figure 1.2).

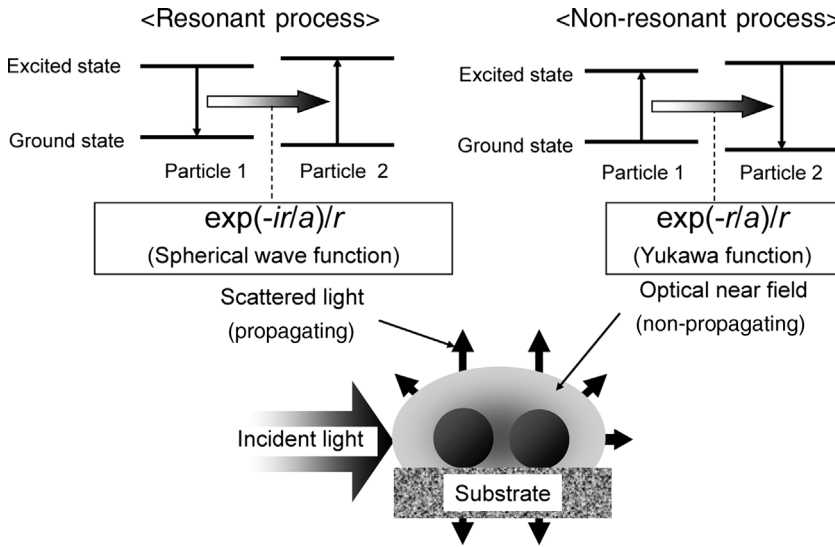


Figure 1.2 Resonant and nonresonant processes.

The interaction in the nonresonant process, corresponding to mediation by the optical near-field, is expressed by a Yukawa function that represents the localization of the optical near-field around the nanometric particles like an electron cloud around an atomic nucleus. Its decay length is equivalent to the material size [4]. On the other hand, the interaction corresponding to the resonant process is expressed by a conventional spherical wave function, and indicates the mediation by a conventional propagating field.

As described above, the optical near-field is an electromagnetic field that mediates the interaction between nanometric particles located in close proximity to each other. Nanophotonics is the technology utilizing this field to realize novel devices, fabrications, and systems. That is, a photonic device with a novel function can be operated by transferring the optical near-field energy between nanometric particles and its subsequent dissipation. In such a device, the optical near-field transfers a signal and carries the information. Novel photonic systems become possible by using these novel photonic devices. Furthermore, if the magnitude of the transferred optical near-field energy is sufficiently large, the structures or conformations of nanometric particles can be modified, which suggests the feasibility of a novel photonic fabrication technique.

With these treatments, a physically intuitive picture of nonpropagating nanometric optical near-fields was established. On reading this, one may understand that the true nature of nanophotonics is to realize ‘qualitative innovation’ in photonic devices, fabrications, and systems by utilizing novel functions and phenomena caused by optical near-field interactions, which are impossible as long as conventional propagating light is used. Nanophotonics undoubtedly has the advantage of exceeding the diffraction limit of light, that is, ‘quantitative innovation’.

This innovation is a really challenging issue in quantum theory because it is the first time the structure, interaction, the temporal evolution, etc. in the region much less than the wavelength, or equivalently those in the region much less than the de Broglie wavelength, has been discussed. This is the first encounter for people to work in such conditions. However, it should be pointed out that this innovation is only a secondary feature of nanophotonics, as compared with the ‘qualitative innovation’. Qualitative innovation has been already realized for devices, fabrications, and systems by appropriately utilizing the true nature of the optical near-field interaction, and some of these are described in the following sections.

1.4 Devices

For nanophotonic device operation, it is essential to transfer the signal and to fix the transferred signal magnitude at the output terminal, which can be achieved by transferring the optical near-field energy and its subsequent dissipation, respectively [20]. Several research groups have recently begun similar discussions on this operation (e.g., refer to [21]). The exciton dynamics of a three-quantum-dot system, as well as a system consisting of a pair of quantum dots, have also been discussed for model nanophotonic functional devices, based on the above formulation and a quantum master equation. The recent analysis includes spin polarization and excitation transfer [22].

As an example, a pair of closely spaced equivalent quantum dots is used as the input terminal of the nanophotonic device (see Figure 1.3). As a result of the optical near-field interaction between the two quantum dots driven by the input optical signal, the quantized energy levels of the exciton in the quantum dots are split into two. One half corresponds to the symmetric state of the exciton, and the other is the antisymmetric state. They represent the respective parallel and antiparallel electric dipole moments induced in the two quantum dots. A third larger quantum dot, located near to the input terminal (see Figure 1.4), is used as the output terminal of the device. The higher energy level of the exciton in this quantum dot is tuned to that of the symmetric state of the input terminal, which is possible by adjusting the size of the third quantum dot. As a result of this tuning, optical near-field energy

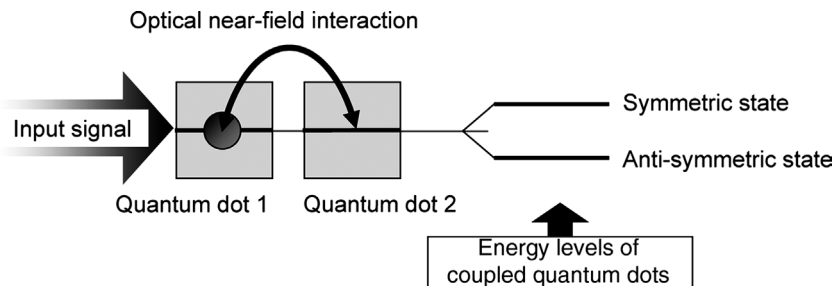


Figure 1.3 The input terminal of a nanophotonic device.

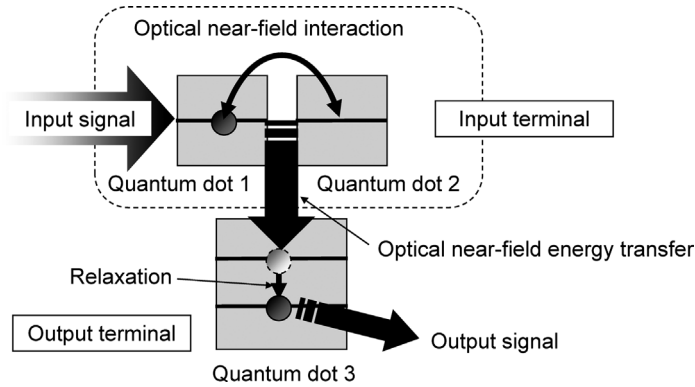


Figure 1.4 Optical near-field energy transfer from the input to the output terminal and subsequent dissipation.

can be transferred from the input to the output terminal, allowing signal transfer. The excitation transferred to the output terminal is dissipated in the third quantum dot immediately by coupling with phonons, which fixes the magnitude of the transferred signal.

A device utilizing the symmetric state of the input terminal in the manner shown in Figure 1.4 is called a ‘phonon-coupled device.’ Examples of phonon-coupled devices include nanophotonic switches [23], logic gates, such as AND and NOT gates [24], content-addressable memory [25], and digital-to-analog converters [26] (see Figures 1.5(a)–(c)). Conversely, a device utilizing the antisymmetric state is called a ‘propagating light-coupled device,’ and examples include an optical buffer memory and super-radiant-type optical pulse generators [27]. They result in qualitative innovative device operations that are impossible when conventional propagating light is used. The nanophotonic switch can be as small as 15 nm. The dynamic behavior of its output signal agrees well with the calculated results based on the dressed-photon model. The figure of merit for this device is 10–100 times larger than those of conventional photonic switches operated using propagating light. The magnitude of the heat dissipated from the nanophotonic switch is estimated to be as low as 10^{-12} W in the case of repetitive 1-GHz operation, which is only 10^{-5} times that of a conventional semiconductor transistor. For the lowest heat dissipation, a device operated using a single photon has also been recently demonstrated [28]. Such a large figure of merit and the ultralow heat dissipation suggest a variety of applications to novel optical computation and information processing systems.

To connect nanophotonic and macroscopic photonic devices, a novel device has been developed that concentrates the propagating light energy to a nanometric region [29] (see Figure 1.6). This device has been named an ‘optical nanofountain’ and its equivalent numerical aperture is as high as 40. Although several plasmonic devices have been developed for this interconnection [30, 31], they cannot realize such a large numerical aperture because the plasmonics method utilizes the classical wave-optical picture. The letters ‘on’ in the word ‘plasmon’ represent the quanta,

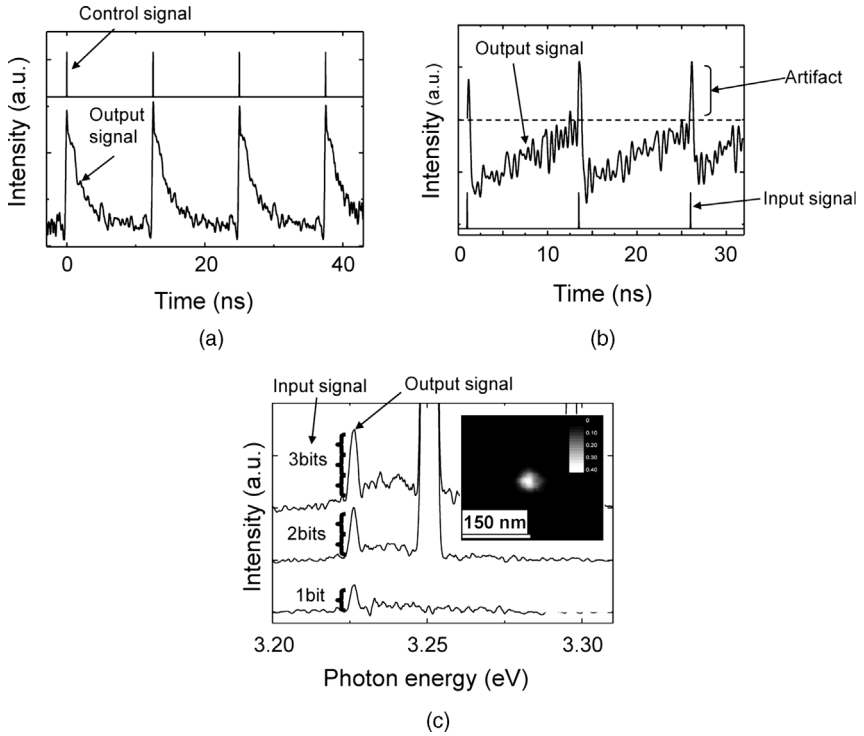


Figure 1.5 The operation of nanophotonic devices. (a) Temporal behavior of the output signal from a nanophotonic switch [23]. (b) Temporal behavior of the output signal from a NOT gate [24]. (c) Spectral intensity of the output signal from a three-bit digital-to-analog converter [26].

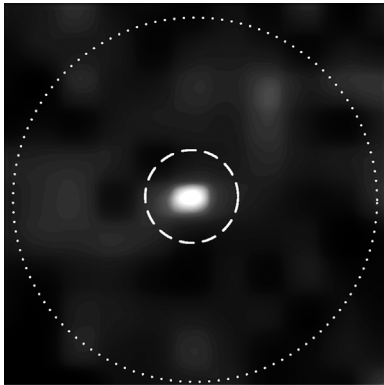


Figure 1.6 Spatial profile of the output signal intensity from an optical nanofountain [29]. The outer circle represents the spot of the incident propagating light.

i.e. the quantum-mechanical picture of the plasma oscillation of free electrons in a metal. Instead of this picture, plasmonics utilizes the classical wave-optical picture by using conventional terminology, such as the refractive index, wave number, and guided mode. Even when a metal is irradiated with light that obeys quantum mechanics, the quantum-mechanical property is lost because the light is converted into the plasma oscillation of electrons, which has a short-phase relaxation time. Therefore, as long as plasma oscillation is used, it is impossible to reduce the device size over the diffraction limit.

1.5 Fabrications

Novel optical nanofabrication techniques beyond the diffraction limit are required for producing a variety of conventional electronic/photonic devices and nanophotonic devices. To fabricate nanophotonic devices, several capabilities are required. For example, a variety of materials must be deposited on a substrate, and the inaccuracy of their sizes and positions must be as low as 1 nm for efficient reproducible optical near-field energy transfer. However, conventional fabrication technologies using electron beams, ion beams, and propagating light cannot meet these requirements due to their low resolution, contamination of and damage to the substrate, and low throughput. To meet the requirements, novel technologies had to be developed, and these have been realized by utilizing optical near-field energy transfer. As representative examples of such nanophotonic fabrication techniques, photochemical vapor deposition and photolithography are described in this section.

Photochemical vapor deposition is a way to deposit materials on a substrate using a photochemical reaction with ultraviolet light that predissociates metalorganic molecules by irradiating gaseous molecules or molecules adsorbed on the substrate. Consequently, the electrons in the molecules are excited to a higher energy level following the Franck–Condon principle. This is an adiabatic process because no molecular vibrations or rotations are excited; that is, the Born–Oppenheimer approximation is valid. After excitation, the electron transits to the dissociative energy level. As a result of this transition, the molecule is dissociated and the atoms forming the molecule are deposited on the substrate. By using an optical near-field as the light source for this deposition scheme, nanometric materials can be fabricated whose size and position are controlled accurately by the spatial distribution of the optical near-field energy. Nanometric metallic particles of Zn and Al and light-emitting semiconductor particles (e.g., ZnO and GaN) have been deposited with size and positional accuracies far beyond the diffraction limit [32–36]. This is an example of a quantitative innovation.

Although ultraviolet light with high photon energy must be used for the above-mentioned adiabatic process, it has been found that an optical near-field with a much lower photon energy (i.e. visible light) can dissociate the molecule. This has been explained by a theoretical model of the virtual exciton–polariton exchange between

a metalorganic molecule and the fiber probe tip used to generate the optical near-field. In other words, this exchange excites not only the electron, but also molecular vibration. This is a nonadiabatic process, which does not follow the Franck–Condon principle, and therefore the Born–Oppenheimer approximation is no longer valid. Several experimental results have been reported, including the nonadiabatic dissociation of diethylzinc (DEZn) molecules by a visible optical near-field and the deposition of nanometric Zn particles [37]. The virtual exciton–polariton model explains this nonadiabatic process quantitatively by introducing the contribution of a phonon excited in the fiber probe tip [38]. A theoretical model for a pseudo-one-dimensional optical near-field probe system has led to a simple understanding of this process [39].

The nonadiabatic process presented above suggests that large, expensive ultraviolet light sources are no longer required, although they have long been indispensable for conventional photochemical vapor deposition. It also suggests that the process can dissociate optically inactive molecules (i.e. inactive to the propagating light), which is advantageous for protecting the environment because most optically inactive molecules are stable chemically and harmless. For example, optically inactive $\text{Zn}(\text{acac})_2$ molecules have been dissociated nonadiabatically using an optical near-field to deposit nanometric Zn particles [40].

Photolithography is a technology used to carve a substrate material. After coating the substrate with a thin film of a photoresist, light is irradiated through a photomask to induce a photochemical reaction in the photoresist. When the aperture on the photomask is smaller than the wavelength of the light, the transmission of the propagating light is sufficiently low, while an optical near-field is generated at the aperture. Using the photochemical reaction between the optical near-field and photoresist, a nanometric pattern beyond the diffraction limit is formed on the photoresist, and a chemical etching process is subsequently used to carve the substrate. Several preliminary experiments have used a fiber probe to generate optical near-fields [41]. Alternatively, a photomask is used to improve the throughput of fabrication dramatically. Practical technologies [42], such as using a two-layered photoresist, have been developed to form deep patterns, thus realizing a quantitative innovation [43, 44].

As long as an adiabatic process is used for photolithography, an ultraviolet light with a high photon energy is required to induce the photochemical reaction via the optical near-field. However, photolithography using a nonadiabatic process is possible, as in the above-mentioned case of photochemical vapor deposition; that is, a photochemical reaction can even be induced using visible light with a very low photon energy. Narrow corrugated patterns were fabricated using nonadiabatic photolithography [45]. This has been analyzed theoretically based on a virtual exciton–polariton model by introducing the contribution of phonons. This result represents a quantitative innovation in photolithography, suggesting that large, expensive ultraviolet light sources are no longer required, and that harmless, chemically stable molecules can be used as the photoresist, even if they are optically inactive. An example of an optically inactive resist film is the one used for electron-beam lithography. A photochemical reaction is induced in this film via a nonadiabatic

process, and fine patterns have been fabricated using a photomask consisting of a two-dimensional array of circular disks [45].

Fabrication using adiabatic processes suffers from the contributions of the low-intensity propagating light transmitted through the aperture on the photomask and of the plasmon generated on the photomask, which limit the resolution of the fabrication. In contrast, since the nonadiabatic process is free of these contributions, greater resolution, higher contrast, and a variety of patterns (e.g., Ts, Ls, and rings) have been realized [45]. A commercial prototype of a compact desktop machine that can fabricate corrugations with a 20–50-nm line width over a 25×25 -mm substrate area has been developed [44].

1.6

Applications to Systems and Evolution to Related Sciences

Nanophotonics has realized quantitative and qualitative innovations. As examples of its evolution to related science and technology, this section describes applications to novel photonic systems and atom photonics. Nanophotonic systems utilize nanophotonic devices. For example, novel architectures have been proposed for optical signal-transmission systems and their performance has been confirmed experimentally. They include interconnections and summation architecture involving an optical nanofountain [25], computing using nanophotonic switches and an optical nanofountain [26], and data broadcasting using multiple nanophotonic switches [46] (see Figure 1.7). They have realized quantitative innovations by decreasing the device size and power consumption beyond the diffraction limit. More importantly, qualitative innovation has been realized because of the novel functions of nanophotonic devices, which are impossible using conventional photonic devices. Quantitative innovation has already been realized by breaking the diffraction limit of optical/magnetic hybrid disk storage density [47].

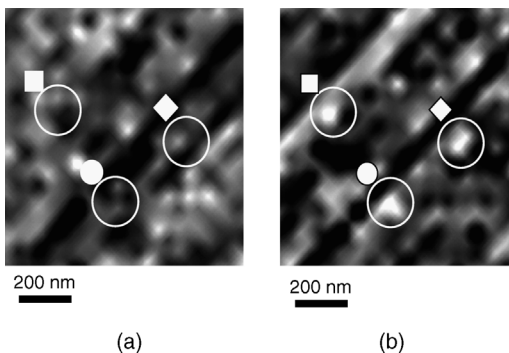


Figure 1.7 The output signal intensity from a data broadcast using three nanophotonic switches [46]. (a) The switches are off. (b) The switches are on.

Qualitative innovation has also been proposed by applying the hierarchy in optical near-fields to memory retrieval [48].

An example of evolution to a related science is atom photonics, which controls the thermal motions of neutral atoms in a vacuum using optical near-fields [5]. Theoretical studies have examined single-atom manipulation based on the virtual exciton–polariton model [19], and experimental studies have involved the first successful guidance of an atom through a hollow optical fiber [49]. Recent studies have examined atom-detecting devices [50], atom deflectors [51], and an atomic funnel [52]. Atom photonics will open up a new field of science that examines the interaction between dressed photons and a single atom.

Basic research for progress in nanophotonics is being actively carried out. An optical near-field problem has been formulated in terms of the Carniglia–Mandel model as a complete and orthogonal set that satisfies the infinite planar boundary conditions between the dielectric and a vacuum. This approach has revealed interesting atomic phenomena occurring near the surface, which have been analyzed based on angular spectrum representation [53, 54]. For example, optical radiation from an excited molecule on the substrate surface has been analyzed, which agreed quantitatively with experimental results [55]. A self-consistent, nonlocal, semiclassical theory on light–matter interactions has been developed to discuss the optical response of a variety of nanostructures [56]. In particular, the size dependence and allowance of a dipole-forbidden transition in a nanometric quantum-dot system were noted [57, 58]. The optical manipulation of nanometric objects in superfluid ^4He has been investigated based on the nonlocal semiclassical theory [59]. Electron transport through molecular bridges connecting nanoscale electrodes has been formulated [60], and a unified method has been proposed to treat extended and polaron-like localized states coupled with molecular vibrations. A one-dimensional molecular bridge made of thiophene molecules has been analyzed numerically. The study of optical near-fields associated with molecular bridges is now in progress. In addition, as basic experimental work, desorption and ionization have been carried out assisted by optical near-fields, and their application to mass spectroscopy has been proposed [61, 62].

1.7

Toward the Future

This chapter reviewed the history and recent progress in nanophotonics, a novel optical technology proposed by the author. It utilizes the local interaction between nanometric particles via optical near-fields. The optical near-fields are the elementary surface excitations on nanometric particles, that is *dressed photons* that carry the material energy. This chapter emphasized that the true nature of nanophotonics is to realize ‘qualitative innovation’ in photonic devices, fabrication techniques, and systems by utilizing novel functions and phenomena caused by optical near-field interactions, which are impossible as long as conventional propagating light is used. Evidence of such innovation was described, that is, novel devices such as switches,

logic gates, and a nanofountain utilizing optical near-fields as a carrier to transmit the signal. Novel fabrications that utilized a nonadiabatic photochemical reaction with visible (not ultraviolet) optical near-fields were also demonstrated, as well as novel nanophotonic information and communication systems that combined nanophotonic devices. Furthermore, it was noted that nanophotonics has evolved into atom photonics.

Nanophotonics is now a key optical technology. However, the name ‘nanophotonics’ is occasionally used for photonic crystals, plasmonics, metamaterials, silicon photonics, and quantum-dot lasers using conventional propagating light. Here, one should consider a stern warning by C. Shannon on the casual use of the term ‘information theory,’ which was a trend in the study of information theory during the 1950s [63]. The term ‘nanophotonics’ has been used in a similar way, although some work in ‘nanophotonics’ is not based on optical near-field interactions. For the true development of nanophotonics, one needs deep physical insight into the *dressed photons* and the nanometric subsystem composed of electrons and photons.

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