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Processing based on fluctuations in a nanometric space and corresponding optical applications in a macroscopic space

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Abstract—Effective utilization of fluctuations in a nanometric physical system is one of the fundamental issues that will be faced in the practical implementation of future processing systems. While fluctuations themselves often show random behavior, it is expected that optimized nanometric structures will be realized by development of a suitable control method on the nanometric scale. On the other hand, recent research on nanophotonics utilizing the characteristic behavior of dressed photons has resulted in several novel applications that work at macroscopic scales. The fundamental principle on which such techniques are based is controlled fluctuations involving interactions between light and materials in a nanometric space, and their corresponding macroscale optical functions. This paper describes concepts, experimental demonstrations, and applications of some recent activity on fluctuation-based nanophotonics, including nanophotonic droplets, dressed photon-assisted giant optical rotation, and compressive near-field optical microscopy.

1. Introduction

Recent research in the field of *beyond von Neumann computing* [1] suggests that effective utilization of fluctuations in a physical system will form an essential part of future processing systems. It is expected that degrees-of-freedom based on fluctuations will be utilized to implement innovative large-scale processing systems. Especially on the nanometric scale, fluctuations in a physical system are one of the most fundamental issues faced in the practical implementation of a processing system. To implement a *fluctuation-based nanometric processing system*, it is essential to develop novel concepts and logic that will replace the conventional ones.

On the other hand, the field of *nanophotonics*, which exploits the local interactions between nanometric particles via optical near fields induced by incident light, has seen rapid progress in recent years, and various studies have been performed [2]. Several of the characteristics of optical near fields can be explained by the behavior of a *dressed photon* (DP), which is a quasi-particle representing the coupled state of a photon and an electron in a nanometric space [3]. A DP excites a multi-mode coherent phonon in a nanometric material, and the DP state is coupled with states of the excited coherent phonon [4, 5, 6, 7]. Because this coupled state can be regarded as an intermediate state during the excitation and relaxation process of the material, multistep excitation and relaxation, and corresponding optical functions, are allowed. Some experimental demonstrations utilizing this phenomenon, called a DP-assisted transition, have been reported, for example, high-yield emission of up-converted optical energy by using organic dye grains [5, 8] and high-intensity emission from indirect transition type semiconductors [6, 7, 9].

The important point in these demonstrations, is that the behavior fundamentally depends on fluctuations of components in a nanometric space, and the optical functions were revealed at a macroscopic scale. This means that precisely controlled fluctuations in a nanometric space can be effectively utilized as characteristic optical functions and processing in a macroscopic space. In this paper, we focus on DPs and related technologies, and we describe concepts and experimental demonstrations on *nanophotonic droplets*, *DP-assisted giant optical rotation*, and *compressive near-field optical microscopy*, which all exhibit macroscopic optical functions based on precisely controlled fluctuations in nanometric spaces.

2. Nanophotonic droplets

We have previously demonstrated a novel technique for autonomous fabrication of a *nanophotonic droplet* (ND) [10, 11, 12], which is a micro-scale spherical polymer structure that contains coupled heterogeneous nanometric components, such as quantum dots (QDs) and organic dye molecules. The sort-selectivity and alignment accuracy of the nanometric components in each ND, as well as the related homogeneity of their optical functions, are due to a characteristic coupling process based on a DP-assisted photo-curing method involving dressed-photon–phonon interactions [13]. The method only requires irradiating a mixture of components with light to induce optical near-field interactions between each component, and subsequent processes based on these interactions. The principle of our method is schematically shown in Fig. 1.

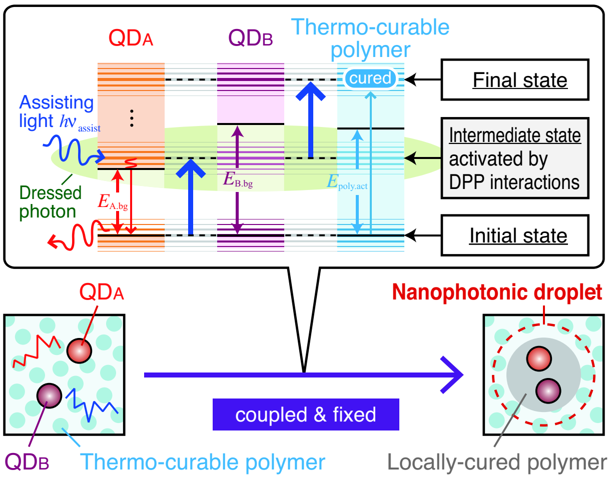

Figure 1: Schematic diagram of process of forming a thermo-curable polymer-based ND via the phonon-assisted photo-curing process.

The ND fabrication process can be induced when energies $E_{\mathrm{A:bg}}$, $E_{\mathrm{B:bg}}$, $E_{\mathrm{poly:act}}$, and $h\nu_{\mathrm{assist}}$ satisfy the condition:

$$E_{\mathrm{A:bg}} < h\nu_{\mathrm{assist}} < E_{\mathrm{poly:act}} < E_{\mathrm{B:bg}}. \quad (1)$$

. Here, if the density is sufficiently high that the QDs can frequently encounter each other, multistep photo-curing occurs due to generation of DPs and corresponding optical near-field interactions between two neighboring QDs, $\mathrm{QD_A}$ and $\mathrm{QD_B}$. As a result, the thermo-curable polymer is locally cured, and the spatial alignment of the QDs that encountered each other is physically fixed by the cured polymer. Because the fabrication of NDs fundamentally depends on components encountering each other due to thermal fluctuations in the mixture, thermal dependency of the process was theoretically and experimentally verified in a previous report by the authors [12].

Figure 2 shows a fluorescence image of the NDs formed using commercially available CdSe-QDs (Sigma-Aldrich, *Lumidots*), CdS-QDs (NN-Labs, *Nanocrystals*), and thermo-curable polymer (Dow Corning Toray, *Sylgard 184*) irradiated with assisting light from a 200 mW laser diode with a wavelength of 457 nm for 30 minutes. As shown, a number of NDs with similar sizes and emission intensities were successfully obtained.

These similarities of massively fabricated NDs are due to the accuracy and homogeneity of their alignment and the combinations of QDs in each ND. Due to the particular structural characteristics of their constituent elements, we have reported an experimental demonstration of effective wavelength conversion based on the novel optical functions of NDs [14]. Because the formation process is induced only when fluctuating heterogeneous QDs encounter each other at the optimum distance to induce appropriate optical near field interactions between the two, their relative spatial positions and corresponding optical functions are autonomously determined. This means that thermal fluctuations of components in the mixture realized optimal nanometric structures, and corresponding optical functions were revealed at a macroscopic scale in the form of light emission from the NDs.

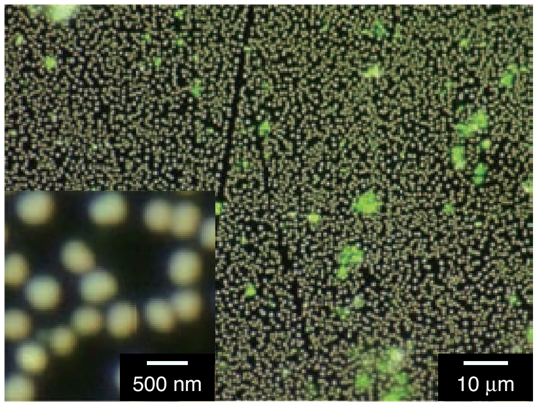

Figure 2: Microscope fluorescence image of densely formed NDs under UV light irradiation and (inset) a magnified view.

3. DP-assisted giant optical rotation

Oxide semiconductors are known to be direct-transition materials with a wide bandgap. Due to their natural abundance, innocuity, and transparency to visible light, they are expected to be widely applied to various optical devices [15]. However, although such oxide semiconductors are highly promising, it is difficult to realize electro- or magneto-induced optical functionalities by general doping methods, because acceptors from the dopants are generally compensated with donors from the numerous oxygen vacancies and interstitial metals in the crystal. On the other hand, according to recent research by the authors' group, a p-type ZnO device, which is one of the most common oxide semiconductors, has been successfully realized by employing annealing using dressed photons [16], a technique known as *DP-assisted annealing*. By using the p-type ZnO device, realization of a p–n homojunction-structured LED that emits at room temperature has been successfully demonstrated. More recently, we applied a voltage to p-type ZnO devices in the in-plane direction instead of the direction perpendicular to the p–n homojunction plane, and experimentally demonstrated novel optical phenomena quite similar to but much larger than the well-known magneto-optical effect. The essential point of that work is is that the distribution of dopants is autonomously optimized by utilizing DP-assisted annealing. DP-assisted annealing controls the fluctuations of dopants in the crystal, causing them to converge to an optimized distribution for inducing DPs and, as a result, corresponding optical functions are exhibited.

In this study, we used an n-type bulk ZnO crystal implanted with N ions (N dopant) serving as a p-type dopant.

A p–n homojunction was formed in the crystal by implanting N ions; however, because this structure was simple, the electrons and holes both exhibited wide spatial distributions, and their recombination probability and the emission intensity were low. Therefore, the devices were subjected to DP-assisted annealing to optimize the dopant distributions; namely, they were annealed with Joule heat by applying a current. During this process, the substrate surface was irradiated with light having a photon energy $h\nu = 3.05$ eV, which is smaller than the bandgap energy of ZnO, $E_g = 3.40$ eV, so as to control the spatial distribution of the N concentration at the p–n junction in a self-organized manner. As a result, DPs were efficiently generated in the N regions, and electrons and holes recombined via these DPs, producing spontaneous emission based on the existence of a good p–n homojunction.

In order to demonstrate giant optical rotation using the p-type ZnO device, the experimental setup shown in Fig. 3 was prepared for observing modulated light among the incident orthogonally polarized light. As shown, a bias current was induced in the device in the in-plane direction instead of the p–n homojunction direction. In such a situation, the magnetic field due to the induced current affected the p–n homojunction. Incident light to the device was temporarily converted to DPs via the dopant, and these DPs interacted with the magnetic field. As the result, corresponding optical modulation was applied to the incident light.

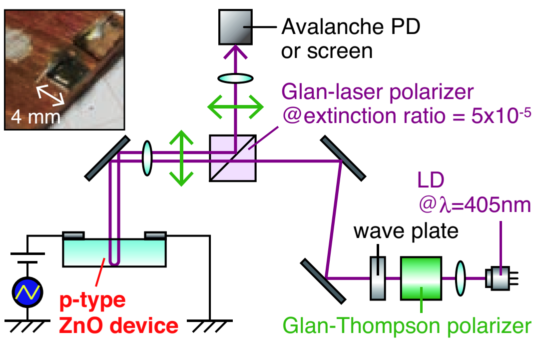

Figure 3: Schematic diagram of experimental setup for measuring optical rotation with orthogonally polarized light. (Inset) Appearance of the prototype p-type ZnO device.

Figure 4 shows the relation between the applied voltage to the device and the polarization rotation. Although the original ZnO crystal does not reveal such large optical rotation of the incident light, extremely large optical rotation was observed in our p-type ZnO device.

The characteristic behavior observed in this research was a result of the optimized distribution of dopants and the corresponding interactions between induced DPs and incident light. What is important is that fluctuations of the dopants due to the Joule heat were effectively controlled by DP-assisted annealing to make the distribution converge to one appropriate for generating DPs and the corresponding op-

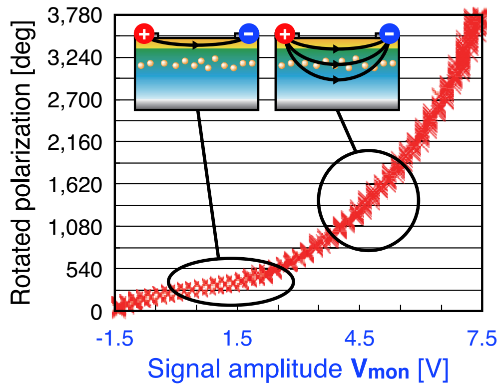

Figure 4: Relation between applied voltage to the device and polarization rotation.

tical functions.

4. Compressive near-field optical microscopy

A *Reader* and a *Device* are fundamental elements of a general authentication system. Recently, the authors developed a novel authentication system based on nanophotonics, what we call a *nanometric artifact-metric system*. Whereas in a conventional system, the Reader reads data from the Device and then their validities are authenticated, in the case of our proposed system, the data to be authenticated is a result of optical near-field interactions between the Reader and the Device in a nanometric space. Because the result of the interactions is strictly dependent on the uniqueness of the physical properties of both the Reader and the Device, security of the system is fundamentally guaranteed. For a simple demonstration of this approach, we previously used a conventional scanning near-field optical microscope (SNOM) as a Reader and a sample containing nanorods, grown by the glancing angle deposition (GLAD) method, as a Device. However, the SNOM required a scanning process, making it quite inconvenient to use the SNOM in a practicable system.

Against such a background, we proposed the concept of a *compressive near-field optical microscope* (CNOM) to demonstrate a practicable nanometric artifact-metric system instead of using an SNOM. A schematic diagram illustrating the concept of the CNOM is shown in Fig. 5.

As shown, the readout results from the probe of the Reader constantly fluctuated based on the feedback setup of NOM, which prevents a collision between the probe and the Device by sensing the shear force between them. This means that the probe constantly reads the result of different scales of interaction during the fluctuation. Because the result of an interaction depends on the element size of the Device at an appropriate scale, which is determined by the readout distance between the probe and the Device, the readout results are expected to contain not only data about a single point but also data about a two-dimensional area in the surroundings. Therefore, we can read out two-

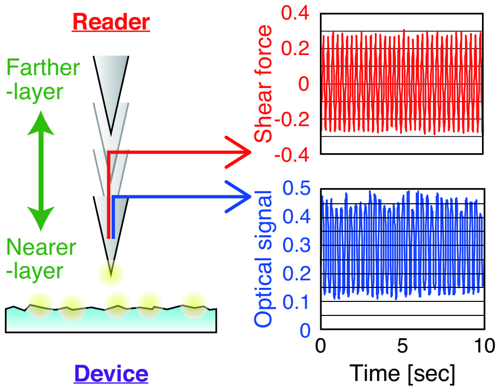

Figure 5: Schematic diagram illustrating concept of a compressive near-field optical microscope (CNOM).

dimensional data without any scanning process by utilizing the fluctuations of the probe. In the case where a basic NOM system is used, readout data from the probe consists of signals due to shear force and the optical response. Here we defined these two types of data as parameter that identifies each Device, and verified their individualities by using different Devices.

For an experimental demonstration, we used the NOM system in an illumination-collection mode with an Au-coated glass fiber probe having a radius of curvature of 50 nm. To control the observation distance, the probe sensitivity to the shear force between the probe tip and the sample was electrically adjusted in multiple steps. A laser diode (LD) with an operating wavelength of 650 nm was used as the light source. Figure 6 shows readout results obtained using two different Devices, Device 1 and Device 2, which consisted of smaller (35 nm diameter) and larger (75 nm diameter) Al nanorods, respectively.

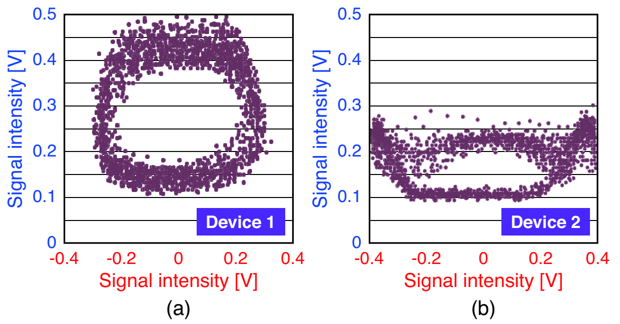

Figure 6: Readout results obtained by using different Devices, which consisted of (a) smaller nanorods and (b) larger nanorods.

As shown, distinct differences were observed between the two. Moreover, because the basic shape of each result surely corresponds to the two-dimensional distribution of nanorods, the characteristic value of each Device is expected to be analogize based on further statistical results, which are obtained without any scanning processes.

5. Summary

We have described some the concepts and experimental demonstrations of some recent activity in research on nanophotonic droplets, dressed photon-assisted giant optical rotation, and compressive near-field optical microscopy. A common point in these systems is that their nanometric mechanisms were optimized by controlled fluctuations of the components based on the behavior of dressed photons, and the results of the optimization were revealed as characteristic optical functions at the macroscopic scale. Focusing on such controlled fluctuations and related technologies is expected to lead to further developments in the research and implementation of future processing systems.

Acknowledgments

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