Basic concepts of nanophotonic devices, nanophotonic systems and nanophotonic fabrication beyond the diffraction limit

Optical near field technology allows one to break the barricades of diffraction light and hence to go beyond the diffraction limit. Thus in reality, optical nearfield technology alone can pave the way for the miniaturization of optical science and technology beyond the diffraction limit, in order to usher in a new era of nanophotonics beyond the diffraction limit. This enables the operation of novel nanophotonic devices. On the basis of such novel nanophotonic devices, a plethora of functions in the form of light source, signal transmission, signal detection, signal control, signal operation, signal interface, etc., are being effectively realized. Moreover, by capitalizing on the transfer of optical excitation from one nanometric system to another via optical near field and also on the hierarchical property of the interactions of the optical near field, one is tempted to consider nanophotonics technology from the point of view of a nanophotonic system. Also, the localization of the dressed photons has paved the way for novel nanophotonic fabrication techniques such as nonadiabatic chemical vapour deposition, photolithography, etc. Basic concepts of nanophotonic devices, nanophotonic systems and nanophotonic fabrication using the principle of optical near field technology are discussed in this lecture.

1 Basic concept of nanophotonic devices

In the optical near field, due to the localized nature of the dressed photon, the long wavelength approximation is not valid. As a result, an exciton can be excited even to an electric-dipole forbiddend energy level. This unusual phenomenon is mainly due to the dressed photon exchange between closely spaced nanoparticles. The above mentioned phenomenon lays the foundation for the design of novel nanophotonic devices that have dimensions far beyond the diffraction limit.

1.1 Prerequisites for nanophotonic device operations

In order to enable the operation of novel nanophtonic devices beyond the diffraction limit on employing optical near field technology, the following prerequisites have to be satisfied:

- Non-resonant exchange of dressed photons in the optical near field between nanometric particles should yield nutation.
- Resonant exchange of free photons should be defected in the far field.
- The resonant energy levles in the far field and the non-resonant energy levels in the near field should be discretized.
- Coupling between two discrete energy levels should yield a symmetric state that is accessible in the far field.
- In the optical near field, coupling between two discrete energy levels should also yield an anti-symmetric state along with the symmetric state.
- When nanomaterial are excited by a far field, only allowed electric dipole energy levels are utilized.

- When nanomaterials are excited by an optical near field, forbidden electric dipole energy levels and allowed electric dipole energy levels are utilized.
- Spatial localization of each nanomaterial should be achieved. Global excitation of nanomaterials as a whole is not needed.
- Spatial information and frequency information of nanomaterial systems should be made use off effectively.

1.2 Principle of operation

To illustrate the principle of operation of a nanophotonic device, a nanometric subsystem consisting of two cubic CuCl quantum dots in a macroscopic *NaCl* crystal is considered. It has been experimentally shown that the translational motion of the exciton center of mass is quantized due to the small exciton Bohr radius for CuCl quantum dots. The given scenario can be attributed to that of a particle confined to a three-dimensional box having an infinite potential barrier. Thus if n_x, n_y and n_z are integer values representing discrete energy levees in a cubic *CuCl* quantum dot with side of length L, the corresponding discrete energy eigen values are given as,

$$E_{n_z,n_y,n_z} = E_b + \frac{(n_x^2 + n_y^2 + n_z^2)\pi^2\hbar^2}{2m(L\text{-}a_b)}, \tag{1}$$

where E_b is the bulk exciton energy, m is the exciton translational mass and a_b is the exciton Bohr radius. When n_x, n_y and n_z have even integer values, the corresponding energy eigen values correspond to electric dipole forbidden energy level states, i.e., they are optically forbidden. But the presence of an effective optical near field allows the participation of such energy levels also. Let $a = L - a_b$. Based on Eq. (1), when the side sizes of the above mentioned quantum dots are of the ration $1 : \sqrt{2}$ respectively, the $(1,1,1)$ exciton energy levels in the small quantum dot depicted as Small QD and the $(2,1,1)$ exciton energy level are resonant with each other. It should be noted that the energy level $(1,1,1)$ is the electric-dipole allowed energy level and $(2,1,1)$ is the electric-dipole forbidden energy level. For the above mentioned resonant condition, the Yukawa function given by $V(r) = \frac{A}{r}\exp(-\mu r)$ yields the required coupling energy for the optical near field interaction. Here, A is the coupling coefficient, μ is effective mass of the Yukawa function and r is the distance between the two quantum dots. Moreover when the small quantum dot is illuminated with an optical signal, an exciton gets excited to the $(1,1,1)$ exciton energy level thereby resulting in a dressed photon. As the long wavelength approximation is not valid, the generated dressed photon excites the exciton to the electric-dipole forbidden energy level $(2,1,1)$ in the big quantum dot depicted as Large QD. From experimental conditions, it is observed that the inter sub level transition time ψ_{sub} from higher exciton energy levels to the lower exciton energy is found to be much faster than the transition time due to optical near field coupling. As a result, the exciton in the $(2,1,1)$ energy level of the big quantum dot undergoes relaxation process to its $(1,1,1)$ energy level. This process guarantees unidirectional signal transfer from the Small QD to the Large QD and is depicted as an animation in Figure 1. This completes the principle of operation of a nanophotonic device under the influence of an optical near field.

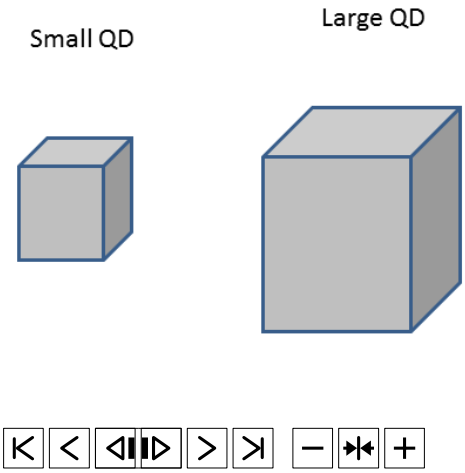

Fig. 1: Unidirectional exciton energy transfer from Small QD to Large QD

2 Basic concept of nanophotonic systems

For novel nanophotonic systems beyond the diffraction limit, in general, even macroscopic free space in vacuum may enact the role of interconnects. Thus any signal can be transferred by the dressed photon exchange between nanometric particles without using any interconnects.

2.1 Unique features of information processing systems

Nanophotonic systems that can operate beyond the diffraction limit posses unique information processing features such as (i) dissipation of low energy, (ii) operation with single photon and (iii) resistance against non-invasive attacks.

2.1.1 Dissipation of low energy

By rule, a nanophotonic system dissipates exciton energy from an electric dipole forbidden energy level in the upper state to an electric dipole allowed energy level in the lower state. If $h(\omega)$ denotes the exciton-phonon coupling energy and $D(\omega)$ denotes the density of states, then the rate of relaxation in a nanophotonic system is given as $\Gamma = 2\pi\hbar^2 |h(\omega)|^2 D(\omega)$.

2.1.2 Operation with a single photon

When more than one excitons are tried to be generated from a nanophotonic device such as CuCl quantum dot, the electric dipole allowed energy level in the lower state tries to still lower its energy that would correspond to the binding energy of the exciton molecule. Due to this, the energy level gets detuned from the input signal and hence the exciton generation no longer takes place. Moreover, the exciton transfer process is a resonant process. It is observed from experiments that if more than one excitons are transferred from one nano device to another, the transfer process is rendered off-resonant and hence

the transfer process does not take place. Moreover, with the energy transfer, a single exciton can remain stable in a given state only if the binding energy of the exciton molecule is large and in such a case, only a single photon gets emitted by a fast relaxation process.

2.1.3 Resistance to non-invasive attacks

Newer and newer innovations in technology are needed in order to cope up with the demands with regard to the information processing capability and the energy conserving capability of various electronic and optical devices. Conventional electronic devices fail miserably to cope up with there demand mainly due to a bottleneck in the switching speed and due to the lack of availability of efficient electrical wires that have to play the role of electrical interconnects for connecting the electronic devices with external devices in order to regulate the direction of the energy flow and also that of the transmitted signal intensity. The outcome of this inefficiency of these electrical interconnects is that a large amount of energy is dissipated in the external macroscopic wires. To add up this already existing woe, the macroscopic electrical interconnects provide feeble security as they are prone to non-invasive attacks. Even though conventional optical devices are far better than the conventional electronic devices with regard to the information processing capability, they experience grave pitfalls with regard to availability of efficient optical interconnects. Conventional optical devices such as optical fibers and dielectric optical waveguides very much need optical interconnects as long as propagating light is used as a signal carrier. Moreover, due to diffraction limit, the size of these optical devices cannot be reduced beyond a certain stage.

The only plausible solution is to resort to optical near field technology which enables one to operate novel nanophotonic systems beyond the diffraction limit. Moreover, for the above mentioned nanophotonic systems, even macroscopic free space in vacuum may enact the role of interconnects. Thus any signal can be transferred by the dressed photon exchange between nanometric particles without using any interconnects. As the signal intensity is decided by the energy dissipation inside the nanometric particles, the possibility of non-invasive attack is completely ruled out.

Based on the above mentioned unique features of a nanophotonic system, prototype demonstration of novel information processing systems are being carried out by the Ohtsu research group. A content addressable memory system depicted in Figure 2 can be thought of as a novel example of a nanophotonic system used for efficient optical routing of data. The input port of such a system is a nanofountain which efficiently handles global summation of digital data and the outport port of the system comprises of nanophotonic switches.

2.2 Applications for information and communications systems

The most important aspect ofa nanophotonic system is that in spite of not having a cental controller in the system, efficient transfer of optical excitations can be realized, which addresses the autonomous behaviour of optical excitations. This can lay the foundation for the realization of a self-organized and distributes complex information and communications technology based systems on internet scale. On using a distributed and autonomous network system, unbalanced traffic load and energy consumptions can be avoided. One can

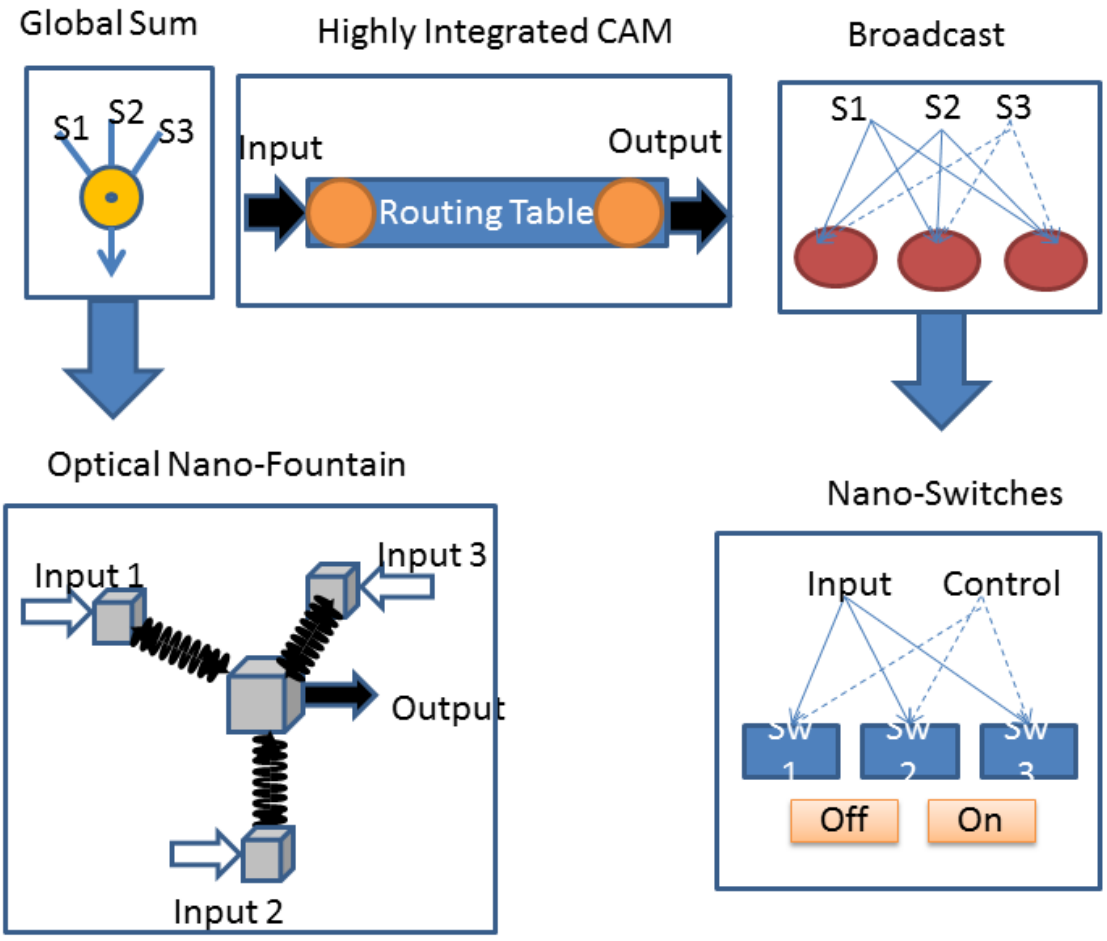

Fig. 2: Content addressable memory of an optical router system

also ensure overall sustainability and reliability as the above mentioned network topology no longer depends on single points of failures.

Moreover as the output signal can be increased due to degraded optical near field interactions robustness against errors can be achieved.

Finally with regard to the energy efficiency of optical excitation transfer, one can say that it is of the order of 10^5 times more efficient than conventional electrical devices. Thus on employing the above mentioned nanophotonics technology, one can easily handle the huge demand in data transfer with more energy efficiently, robustness, overall sustainability and reliability.

3 Basic concept of nanophotonic nanofabrication

The basic concept of nanophotonic fabrication beyond the diffraction limit can be best explained on the basis of the exciton-phonon polariton model which emphasises that the localization of the dressed photons are due to the coherent phonons.

3.1 Exciton-phonon polariton model

The exciton-phonon polariton model is basically used for investigating the physical mechanism of near field optical chemical vapour deposition. According to the model, the exciton-phonon polaritons are generated at the apex of an optical near field probe. When gas and adsorbed molecules are placed very close to the optical near field probe tip, the exciton-phonon polaritons are transferred from the probe tip to the adsorbed gas molecules. The dressed photons incident into the probe-tip are responsible for the electronic excitations near the probe tip. These electronic excitations inturn result in the anharmonic coupling of the phonons, thereby forming a renormalized phonon. Hence multiple phonons can be thought of as coherent phonons in the original representation which can interact simultaneously with an exciton or an exciton polariton.

3.2 Localization of dressed photons

A relevant nanometric system can be considered in the form of an optical near field probe where light interacts on a nanometric scale with both phonons and electrons in the probe. This system can be perceived as a one-dimensional atomic or molecular chain that is coupled with photon and phonon fields. Thus the molecular chain is composed of finite number of molecules. Each molecule present in the molecular chain is called a molecular site. The separation between two consecutive molecular sites is a characteristic scale of the optical near field system. Due to the short range interaction nature of the optical near fields, the dressed photons can hop to the localization of dressed photons. The localization of the dressed photons is mainly due to the presence of coherent phonons. This can be explained as follows: when the photons and phonons do not interact with each other, the photons are symmetrically spatially distributed. This means that the localization of photons do not take place at any specific site, as the photons spread over the entire nanometric system as a result of photon hopping. However, when impurities are added to the nanometric system, during doping process. Instead of spreading or delocalizing to the entire nanometric system, the photons move from one impurity site to another impurity site. When the photon-phonon coupling becomes very large, the photons are forced to stay in the initial impurity site instead of hopping and thereby become localized. Thus the coherent state of the phonons form the dressed photons and are responsible for their localization.

3.3 Principle of adiabatic near field optical chemical vapour deposition

Optical near field technology, is instrumental in the fabrication of nanostructures precisely controlling their size and position by way of adiabatic chemical vapour deposition. The optical near field initiates a two-stage process, namely photo dissociation and adsorption, for achieving adiabatic chemical vapour deposition. This is mainly because of the localization of the photons that are dressed by the coherent phonon states. These localized dressed photons are present at the tip of the optical near field probe. When gas molecules or adsorbed molecules are placed very close to the probe tip, the dressed photons are transferred to them and as a result, the molecules undergo vibrational excitation. This further leads to the electronic excitation. This culminates with the dissociation of the molecules. Hence

using an optical near field, phonon assisted photo dissociation of molecules take place. This satisfies the Franck-Condon principle for adiabatic chemical vapour deposition which states that for photo dissociation to occur, the gas molecules should be excited from the ground state to an excited electronic state by resonating the reacting molecular gases by the conventional propagating light. After relaxing to the dissociation channel, the dissociated atoms finally get adsorbed to the surface of the substrate.

Conventional photochemical vapour deposition thus involves the principle of adiabatic nanofabrication where propagating light resonates the absorption band of metal-organic vapour. An important point to note is that the photon energy of the light source should be much greater than that of dissociation energy. The entire procedure of the conventional photochemical vapour deposition can be subdivided into two halves:

1. One that involves gas-phase photo dissociation where the molecules are excited from the ground state to the excited electronic state by the resonant photons;

2. After the excited molecules undergo relaxation process and go to the dissociation channel, they constitute as dissociated metallic atoms. Finally, they get adsorbed to the substrate

3.4 Principle of non adiabatic near field optical chemical vapour deposition

The principle of non-adiabatic photochemical vapour deposition can be best explained on the basis of an optical near field. Due to optical near field, electric-dipole forbidden energy transitions also occur, which enhances photo dissociation process by making possible four mechanisms such as (a) two-photon absorption process, (b) transition to the intermediate energy level, (c) transition to an excited state of a molecular vibration mode and (d) direct transition from the singlet ground state to the dissociative triplet state.

In this case, the Frank-Condon principle is violate such that during the non resonant condition of electronic transition, a photo dissociation process is observed. The non resonant condition is because of the homogeneous distribution of the conventional propagating light energy in a molecule. As a result, the electric field of the propagating light are able to excite only the electrons in the molecule and not the molecular vibration. The molecular vibration mode is excited only because of the steep spatial gradient of the optical near field, thereby rendering optical near field energy distribution spatially inhomogeneous. As a result, even in the non resonant condition, due to the changes in the molecular orbital, excitation of the molecular vibration modes take place. The electronic response in this case becomes inhomogeneous and the molecules get polarized.

3.5 Principle of non adiabatic photolithography

The concept of non adiabatic photolithography can be explained on the basis of the dressed photon-coherent phonon model. Using a visible light source, commercial photoresists can be patterned. The propagating light does not pattern the photoresist as the photoresist is sensitive only to UV propagating light. However a dressed-photon coherent phonon gets generated at the photoresist edge. The photoresist gets activated by the transfer of energy

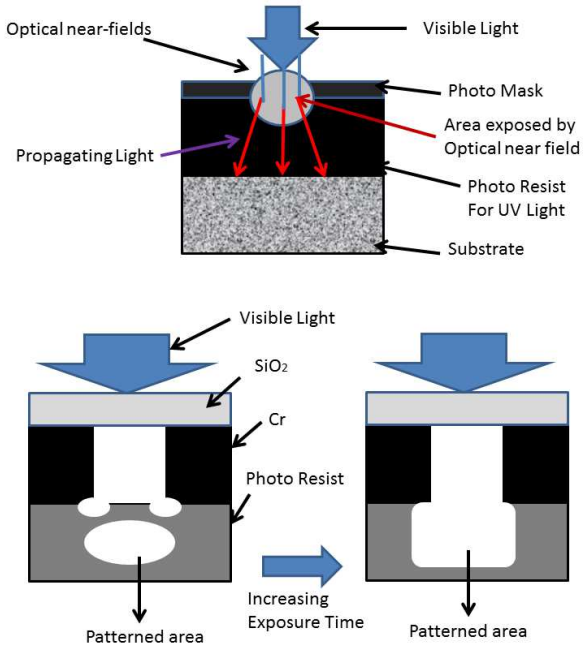

Fig. 3: Phonon assisted non adiabatic photolithography

from the dressed photon coherent phonon to the photoresist and thereby gets patterned due to the phonon assisted process. Moreover, as the energy gets transferred not only to the surface of the photoresist but also to its interior, the photoresist gets effectively patterned within a short exposure time. Thus by properly manipulating the exposure time, the photoresist can be patterned to have a stable spatial profile. By employing a light source of an appropriate frequency, high resolution can be achieved when the wavelength of the light source is greater than the wavelength of absorption band edge of the photoresist. Hence phonon assisted photolithography is not expensive as it does not require either short wavelength X-ray or UV light source for patterning. The advantage of using phonon assisted photolithography is that complicated patterns can be obtained with high resolution when subject to multiple exposures as the photoresist is insensitive to incident visible light. Phonon assisted photolithography has the ability to pattern even an optically inactive film. Phonon assisted non adiabatic photolithography is portrayed in Figure 3

4 References and additional reading

1. Motoichi Ohtsu, Kiyoshi Kobayashi, Tadashi Kawazoe, Takashi Yatsui and Makoto Naruse, *Principles of Nanophotonics*, CRC Press, Taylor and Francis, NewYork, 2008 (Chapters 4 and 5).

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