Laser-Guided Assembly of Nanosystems

NSF NIRT Grant 0404030
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The promise of nanotechnology won't be realized unless nanometer-scale structures can be assembled together inexpensively into a working system. The goal of this work is to develop and test a revolutionary tool that uses light-pressure forces to rapidly assemble complex nanosystems comprised of structures ranging in size from ~10nm to 1μm. We plan to develop a tool to assemble a nanosystem layer-by-layer using light pressure forces to produce multiple, independent optical traps for organizing simultaneously thousands of nanometer-scale structures within each layer. The optical traps will be produced either by rapidly scanning a laser beam from one trap location to the next, relying on the viscosity of the medium to stabilize the position until the trap is refreshed, or by generating a hologram, where multiple optical traps are created simultaneously by controlling the intensity and phase profile of the beam using a spatial light modulator. Either way, the tool will have to compensate in real-time for the scattering environment of the trap during the layer-by-layer assembly. ***Therefore, there are two elements at the core of this proposal: 1. the efficient simulation of the dynamic electromagnetic environment of the trap, which is used to predict in real-time the required intensity and phase profiles for the laser; and 2. the concomitant synthesis through adaptive optics of the trap.***

***Experimental*:**

1. We have explored the use of light-pressure forces (optical tweezers) for rapidly assembling 2D and 3D microarrays of living cells to control and test the effect of the microenvironment on cell development and biofilm formation and accurately predict in vivo responses. In pioneering work two decades ago, Ashkin and co-workers[1] demonstrated the utility of a single beam, gradient force optical trap for capturing and holding bacteria, and manipulating single cells and organelles. Right now, we are investigating the prospects for using this same toolset to organize more complex systems with nanometer precision is avidly being explored. For example, we

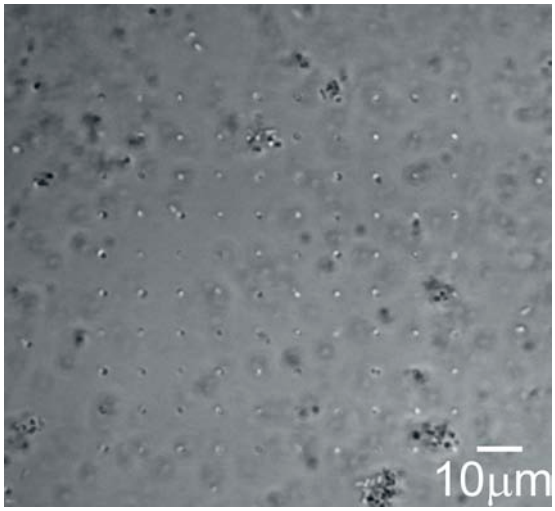

Figure 1. 10x10 Living cell microarray P. aeruginosa bacteria formed using a time-shared array of optical tweezers.

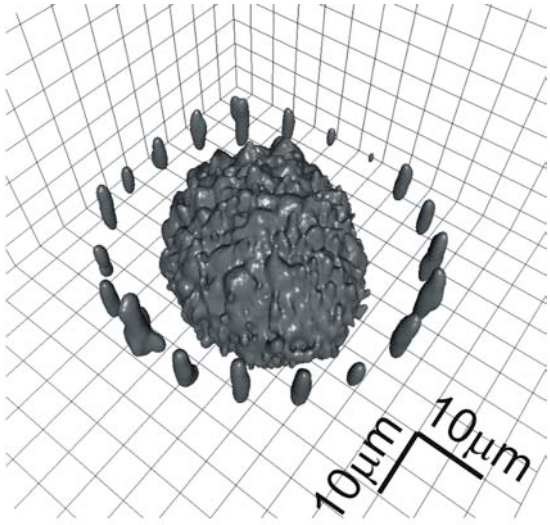

Figure 2. Heterotypic microarray of Swiss-3T3 mouse fibroblast surrounded by a ring of 20 P. aeruginosa bacteria formed with optical tweezers.

have leverage these same principles to assemble living cells into 2 and 3D microarrays by using multiple, independent optical traps to organize hundreds to thousands of cells within a layer. The feasibility of using multiple optical traps to manipulate cells has already been demonstrated in preliminary work: Figure 1 shows a 10x10 living cell micro array bacteria, P. aeruginosa configured using two acousto-optic deflectors (AODs). Moreover, the cells can be positioned in 3D using a holographic array of traps formed using a Spatial Light Modulator (SLM) in conjunction with AODs as demonstrated in Figure 2. By juxtaposing two diffractive optical elements, an AOD with an SLM, we ultimately plan to develop a tool that can produce a 3D array comprised of >1,000 cells or nanoparticles.

2. Furthermore, we have encapsulated the living cell microarrays within a hydrogel to maintain the local 3D microenvironment. Hydrogels are especially efficacious for this application because of their demonstrated biocompatibility. They are hydrophilic and have a high water content; they are pliable; and they allow transport of nutrients to the cell and waste away from it. Hyrdogels have already been used to immobilize various cell types that can attach, proliferate and produce matrix within millimeter-scale structures *without loss of cell viability*. The Poly(ethylene glycol) diacrylate (PEGDA)-based hydrogels are formed in the presence of the arrayed cells by photo-crosslinking. The PEGDA covalently crosslinks via radical chemistry upon exposure to UV (λ=365nm) light. We have demonstrated our capacity for assembling heterogenous microarrays of cells in hydrogel as shown in Figure 2: Pseudomonas aeruginosa is configured into a 2D microarray surround and Swiss 3T3 cell into a using a time-shared holographic optical trap.

3. We have prepared through self-assembly silica colloidal crystals and 75nm diameter ZnS nanoparticles with an effective refractive index of ~1.8 to be used together to form a photonic integrated circuit. We plan to introduce with nanometer-scale precision ZnS nanoparticles at strategic locations in the silica photonic crystal to form complex waveguide structures. To demonstrate feasibility, we have already trapped simple arrays of 90nm ZnS nanoparticles in a unique solvent system as illustrated in Figure 5. We designed the solvent system to satisfy all the important criteria for trapping of nanoparticles inside of a self-assembled colloidal crystal. Successful permanent trapping of nanoparticles in colloidal crystals requires the refractive index of the solvent to match that of the colloidal crystal to eliminate undesirable scattering, the nanoparticles to not aggregate, and the solvent to be gelled to lock the nanoparticles into place once they have been moved to their proper location. The solvent system we have developed consists of a water, glycerol, monomer, cross-linker, and photoinitiator mixture. The ratio of glycerol to water is selected to index match the silica based colloidal crystal; the monomer can be polymerized by the application of UV-light, gelling the fluid and locking the nanoparticles into place.

***<u>Theoretical</u>*:**

Our efforts this past year focused on the development of a field solver that utilizes the generalized field propagator concept to provide for the computationally efficient update of the field solution due to an incremental change in the scattering topography (i.e., material properties). The concept of the generalized field propagator is captured by the following statement of the linear electromagnetic boundary value problem of interest

$$\mathbf{E}^{new} = \mathbf{E}^{old} + \mathbf{G}^{new} \cdot \varepsilon_{change} \mathbf{E}^{old}$$
$$\mathbf{G}^{new} = \mathbf{G}^{old} + \mathbf{G}^{old} \cdot \varepsilon_{change} \mathbf{G}^{new} \quad (1)$$

The second equation is Dyson's equation and provides a systematic means for the construction of the Green's tensor for the new environment, $\mathbf{G}^{new}$, in terms of the change in the environment, ε_{change}, and the Green's tensor prior to the change, $\mathbf{G}^{old}$. Once $\mathbf{G}^{new}$ is available, the new electric field, $\mathbf{E}^{new}$, in the region where the change occurred is obtained from the first equation in terms of the electric field $\mathbf{E}^{old}$ that existed prior to the change in the electromagnetic properties of the environment. Subsequently, the calculation of the new field anywhere in the environment of interest can be performed very efficiently using the Lippmann-Schwinger equation

$$\mathbf{E}^{new} = \mathbf{E}^{old} + \mathbf{G}^{old} \varepsilon_{change} \mathbf{E}^{new} \qquad (2)$$

Thus, for example, in the case of scattering by a single particle in a homogeneous medium, the starting Green's tensor is the homogenous medium Green's tensor (which is known in closed form), and the starting electric field is the electric field of the incident beam in the absence of the particle. At the end of the computation described by (1), the resulting Green's tensor, $\mathbf{G}^{(1)}$, accounts for the presence of the particle and the resulting electric field, $\mathbf{E}^{(1)}$, is the superposition of the incident and scattered fields. If the change in the scattered field due to the introduction of a second particle is desired, the calculation commences from this point, using the calculated $\mathbf{G}^{(1)}$ and $\mathbf{E}^{(1)}$ as the "old" Green's tensor and electric (or incident) electric field, respectively.

We have implemented this process successfully for the case of particles in homogeneous media. Since Dyson's equation is an integral equation, the major bulk of the computational complexity of the process described by the system in (1) is associated with its solution. Our initial implementation relies upon the brute force solution of the integral equation. Its computational efficiency is acceptable for handling several tens or a few hundreds of electrically small particles. However, for handling several hundreds or thousands of particles, fast methods must be utilized for its solution. The implementation of fast solvers for the solution of (1) will be undertaken during the second year of this effort.

For the case where optical manipulation of particles is performed in the presence of a layered stratified medium, the reflective/scattering properties of the background medium must be taken into account in the development of the Green's tensor. For the case of planar stratified media, the elements of the electrodynamic Green's tensor are in terms of Sommerfeld integrals that require structure-specific manipulation of the integrand to expedite their numerical computation. Furthermore, their integral form hinders the direct utilization of fast integral equation solution (both fast multipole-based and FFT-based) methods for expediting the solution of light scattering by clusters of particles in the presence of a stratified substrate.

To address this issue we developed a systematic methodology for the accurate approximation of the Sommerfeld integrals in terms of a finite sum of cylindrical and spherical waves. Our methodology has the unique attribute that it is applicable to stratified media of arbitrary composition and that it exhibits high accuracy both in the near field zone (particularly, in the immediate vicinity of the source) and far away from the source. More importantly, the cylindrical waves (Hankel functions) and the spherical waves (spherical Hankel functions) involved in its expression are, respectively, the Green's functions for the two-dimensional and the three-dimensional scalar Helmholtz equations in homogenous media. Thus, the well-understood computational framework of fast integral equation solvers in homogeneous media can be readily utilized for our purposes.

References

[1] A. Ashkin and J.M. Dziedzic, Appl Phys. Lett. 19 (1971) 283; and A. Ashkin and J.M. Dziedzic, *Optical trapping and manipulation of single living cells using infra-red laser beams,* Ber. Bunsen-Ges. Phys. Chem., 98, (1989) 254-260.