REP	ORT DOCU	MENTATI	ON PAGE		For	m Ap	pproved OMB NO. 0704-0188	
The public reposed searching exist regarding this Headquarters S Respondents sl of information if PLEASE DO NO	orting burden for the ing data sources, g burden estimate of Services, Directora hould be aware tha it does not display DT RETURN YOUF	his collection of in gathering and mair or any other aspe ate for Information t notwithstanding a a currently valid OI R FORM TO THE A	formation is estimated to taining the data needed, ct of this collection of i Operations and Repor iny other provision of law MB control number. BOVE ADDRESS.	average and conformat ts, 121 , no per	ge 1 hour pe ompleting and tion, including 5 Jefferson tson shall be	er res d revie g sug Davis subjee	ponse, including the time for reviewing instructions, ewing the collection of information. Send comments gesstions for reducing this burden, to Washington Highway, Suite 1204, Arlington VA, 22202-4302. ct to any oenalty for failing to comply with a collection	
1 REPORTI	DATE (DD-MM	YYYY)	2 REPORT TYPE				3 DATES COVERED (From - To)	
07_02_2017	7	)	Einal Report				10-Sep-2009 - 9-Nov-2016	
	ID SUDTITI E				5. 00			
4. IIILE AI		· · · · · · · · · · · · · · · · · · ·	1 D 6 6 6		Sa. CO	JNH	RACI NUMBER	
Final Repo	II. LIGHT-ASSIS	led Assembly	and Reconfiguration		W91	INF-	-09-1-04/3	
Complex C	plical Materia	us using Micro	opnotonic Tempiat	es	5b. Gl	RAN	T NUMBER	
					5c. PR	OGF	RAM ELEMENT NUMBER	
					6111	02		
6. AUTHOR	S				5d. PR	5d_PROJECT NUMBER		
Michelle L	Povinelli							
					5e. TA	SK 1	NUMBER	
					5f. W0	ORK	UNIT NUMBER	
7. PERFOR	MING ORGANI	ZATION NAMI	ES AND ADDRESSE	S		8.	PERFORMING ORGANIZATION REPORT	
University of	of Southern Calif	òrnia				NU	JMBER	
Contracts &	Grants							
3720 S. Flo	wer St.		0 0501					
Los Angele	s, CA	9008	9 -0701					
9. SPONSO (ES)	RING/MONITO	RING AGENCY	NAME(S) AND ADI	DRESS	5	10. A	SPONSOR/MONITOR'S ACRONYM(S) Aro	
U.S. Army F	Research Office					11.	SPONSOR/MONITOR'S REPORT	
P.O. Box 12211 Descent Triangle Dark NG 27700 2211				NUMBER(S)				
Research II	langle Park, NC	27709-2211				568	301-MS-PCS.23	
12. DISTRIE	UTION AVAIL	IBILITY STATE	EMENT					
Approved for	Public Release;		Imited					
The views, of the Army	MENTARY NO pinions and/or fin position, policy o	ndings contained or decision, unles	in this report are those s so designated by oth	e of the er docu	e author(s) a umentation.	nd sł	nould not contrued as an official Department	
14. ABSTRA	ACT							
In this wor	<ul> <li>we successf</li> </ul>	ully proposed	and demonstrated	a new	v method	for n	naking ordered arrays of nanoparticles	
The technic	ue uses a tem	plate made of	silicon with small	holes	in it. Shi	ning	laser light at the template creates a	
strong opti	cal field in the	holes: these t	hen serve as optica	1 traps	s. By desi	gnin	ig the template appropriately, we can	
assemble o	rdered arrays	of dielectric of	r gold nanoparticle	s Cha	anging the	e lase	er wavelength allows the pattern of	
traps, and t	hus particles,	to be reconfig	ured.	5. Ciii		14.5	er waverengen and we une pattern er	
15. SUBJEC	CT TERMS							
self assembl	y, photonic cryst	als, optical force	s, optical trapping, gol	d nanoj	particles			
16 00000					15 3113 57		10. NAME OF DESDONGIDUE DEDGON	
16. SECURI	I Y CLASSIFICA	ATION OF:	- $ABSTRACT$	OF	ID. NUME	SEK	19a. NAME OF KESPONSIBLE PEKSON Michelle Povinelli	
a. KEPORT	D. ABSIKACT	C. THIS PAGE				,	19b TELEPHONE NUMBER	
	00						+12-137-4086	

### **Report Title**

Final Report: Light-assisted Assembly and Reconfiguration of Complex Optical Materials using Microphotonic Templates

### ABSTRACT

In this work, we successfully proposed and demonstrated a new method for making ordered arrays of nanoparticles. The technique uses a template made of silicon with small holes in it. Shining laser light at the template creates a strong optical field in the holes; these then serve as optical traps. By designing the template appropriately, we can assemble ordered arrays of dielectric or gold nanoparticles. Changing the laser wavelength allows the pattern of traps, and thus particles, to be reconfigured.

# Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received	Paper
10/24/2016	21 Shao-Hua Wu, Ningfeng Huang, Eric Jaquay, Michelle L. Povinelli. Near-Field, On-Chip Optical Brownian Ratchets, Nano Letters, (): 5261. doi:
10/24/2016	22 Aravind Krishnan, Ningfeng Huang, Shao-Hua Wu, Luis Javier Martínez, Michelle L. Povinelli. Enhanced and selective optical trapping in a slot-graphite photonic crystal, Optics Express, (): 23271. doi:
TOTAL:	2

### Number of Papers published in peer-reviewed journals:

### (b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

### Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper
09/01/2012	5 Camilo Mejia, Eric Jaquay, Luis Martinez, Avik Dutt, Michelle Povinelli. Light-assisted templated self assembly using photonic crystal slabs, Optics + Photonics, Optical Trapping and Micromanipulation VIII. 22-AUG-11, . : ,
TOTAL:	1

### Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

### **Peer-Reviewed Conference Proceeding publications (other than abstracts):**

Received	Paper
----------	-------

- 07/31/2014 14.00 LJ Martinez, E Jaquay, Jing Ma, ML Povinelli. Fabrication and optical characterization of high-Q guided mode resonances in a graphite-lattice photonic crystal slab, Conference on Lasers and Electro-Optics (CLEO). 06-MAY-12, . : ,
- 08/20/2013 8.00 Ningfeng Huang, Eric Jaquay, Chenxi Lin, Jing Ma, Luis J. Martinez, Camilo A. Mejia, Michelle L. Povinelli. Guided resonance modes in photovoltaics and light-assisted self assembly, THE FIFTH INTERNATIONAL WORKSHOP ON THEORETICAL AND COMPUTATIONAL NANO-PHOTONICS: TaCoNa-Photonics 2012. 24-OCT-12, Bad Honnef, Germany. : ,
- 08/31/2015 17.00 Eric Jaquay, L. J. Martinez, C. A. Mejia, M. L. Povinelli. Experimental demonstration of light-assisted, templated self assembly using photonic-crystal slabs, Conference on Lasers and Electro-Optics. 09-JUN-13, . : ,
- 08/31/2015 18.00 Ningfeng Huang, Luis Javier Martinez, Eric Jaquay, Camilo A. Mejia, Debarghya Sarkar, Michelle L. Povinelli. Light-assisted Templated Self-Assembly of a GoldNanoparticle Array, Conference on Lasers and Electro-Optics. 06-JUL-14, . : ,

TOTAL: 4

		(d) Manuscripts
Received	<u>Paper</u>	
TOTAL:		
Number of Mar	nuscripts:	
		Books
Received	<u>Book</u>	
TOTAL:		
<u>Received</u>	Book Chapter	
TOTAL:		
	H	Patents Submitted
		Patents Awarded
		Awards

NAME	PERCENT_SUPPORTED	Discipline
Aravind Krishnan	0.21	
Ahmed Morsy	0.21	
Shao-Hua Wu	0.14	
FTE Equivalent:	0.56	
Total Number:	3	

NAME

PERCENT SUPPORTED

### FTE Equivalent: Total Number:

### Names of Faculty Supported

NAME

PERCENT\_SUPPORTED

FTE Equivalent: Total Number:

### Names of Under Graduate students supported

NAME

PERCENT\_SUPPORTED

FTE Equivalent: Total Number:

# Student Metrics This section only applies to graduating undergraduates supported by this agreement in this reporting period The number of undergraduates funded by this agreement who graduated during this period: ...... The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:..... The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:..... Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):..... Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:..... The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense ...... The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense ...... The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: ..... The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields: ..... </

NAME

**Total Number:** 

<u>NAME</u>

**Total Number:** 

### Names of other research staff

<u>NAME</u>

PERCENT\_SUPPORTED

FTE Equivalent: Total Number:

Sub Contractors (DD882)

**Inventions (DD882)** 

**Scientific Progress** 

**Technology Transfer** 

# Light-Assisted Assembly and Reconfiguration of Complex Optical Materials Using Microphotonic Templates

Michelle L. Povinelli, University of Southern California

### Statement of the problem studied

Self-assembly methods [1] provide a widely used "bottom-up route" to making complex materials. Self assembly of colloidal particles has proven useful for making nanostructured, photonic materials such as photonic crystals [2-4] and metamaterials [6]. However, a fundamental limitation of self assembly is that only certain, energetically favorable structures are formed. To expand the range of particle patterns that can be assembled, templates can be used to guide particles into their desired locations [7-10]. However, the pattern of particles assembled in this way is static, and not easily reconfigured. Methods for on-demand assembly and reconfiguration of the particle pattern would greatly expand the range of potential applications for complex photonic materials formed by assembled nanoparticles.

In this work, we introduced a method that uses light to "assist," or drive, the assembly process [11] (Figure 1). Our technique uses laser light incident on a patterned template to create an array of strong optical traps, which pull particles into place from solution. We studied the types of particles that could be assembled in this manner, and what patterns could be formed. We further examined how

interactions between particles affect the possible patterns and their stability. The use of incident light to control assembly also provides a route to reconfiguration: theoretically, we have identified a mechanism for changing the assembled particle pattern via tuning of the wavelength of polarization of the incident laser [11]. We further assessed the ability of this assembly method to serve as the basis for (i) a nanoparticle size sorter and (ii) rectified Brownian motion, or "ratchets".

## Summary of the most important results



**Figure 1: Light-assisted, templated self assembly.** (left) Light is incident from below on a template, formed by a silicon photonic crystal slab. The slab enhances the light intensity near the holes, creating an array of optical traps for nearby particles in solution. (right) Microscope image of assembled gold particles with 200nm diameter, taken from our Ref. [5].

**Assembled dielectric nanoparticle arrays:** We demonstrated the assembly of over 100 dielectric particles in a square lattice [12]. Figure 2 shows snapshots of the process. The photonic crystal template is visible in the background of each frame. When the laser beam is turned on, nanoparticles are attracted toward the slab, and begin to occupy the sites of the square lattice (Figure 2(a)). As time progresses, additional particles diffuse into the region where the intensity is high and begin to form a cluster (Figure 2(b)). Eventually, a regular array is formed (Figure 2(c)). The square symmetry of the particle arrangement is evident from the picture. When the beam is turned off, particles immediately diffuse away from the slab via Brownian motion (Figure 2(d)). These frames were recorded with a relatively dilute solution of particles for clarity of imaging, and represent an elapsed time of

approximately one hour. We have verified that faster cluster formation occurs with higher concentrations, or by using particle flow, for assembly times of a few minutes.

Each site of the square lattice may be viewed as an optical trap. We obtained trap stiffness values comparable to those reported for single-particle, microphotonic near-field traps [13], which are an order of magnitude higher than standard optical tweezers. For polystyrene particles, particle interactions appeared to be minimal.



**Assembled gold nanoparticle arrays:** For gold nanoparticles, we observed strong interparticle interactions. Left unchecked, these can inhibit the formation of regular arrays. In our experimental work, we observed that for lattices with square symmetry, the gold nanoparticles formed chains [14]. We formulated a theoretical approach to predict the effect of interactions on the assembled pattern. Using this model, we identified a template with hexagonal symmetry that would promote the assembly of regular arrays and observed the assembly experimentally (Figure 1b) [5]. We found that in this case, the interparticle interactions act to stabilize the array, creating highly stable assemblies.

**Pattern reconfiguration:** Different templates support different optical field patterns, and thus trap particles in different locations. We explored and demonstrated this concept in our early simulation paper, Ref. [11]. More recently, we refined our template designs to be fully compatible with our experimental infrastructure (Figure 3).

**Nanoparticle sorting:** In the course of our work, we designed templates that reduce the optical power required for optical trapping. These templates tightly concentrate light in thin slots, boosting the optical trapping strength for fixed laser power [15]. We fabricated and characterized these templates [16] and used them for assembly experiments. We found that particles could be selectively trapped based on size [17], suggesting a mechanism for sorting nanoparticles out of mixtures.

*Optical ratchets:* Study of classical ratchet potentials has shown that for a particle subject to Brownian motion and confined to a asymmetric potential, switching the potential on and off can make the particle move in one direction [18]. The transport rate depends on particle size, leading to techniques for sorting microscale particles and DNA [19, 20]. Optical ratchets have previously been implemented using fluctuating optical lattices [21], but never with integrated microphotonic devices. We have reported the *first integrated photonic ratchet* in *Nano Letters* [22], using a modified version of our assembly method.



We designed a template with spatial asymmetry and calculated the resulting optical potential. We showed experimentally that modulating the incident light rectifies the Brownian motion of assembled nanoparticles, preferentially driving them in one direction.

### Bibliography

- 1. G.M. Whitesides and B. Grzybowski, Self-assembly at all scales. *Science*. **295**: 2418-2421 (2002).
- 2. Y. Xia, B. Gates and Z.-Y. Li, Self-assembly approaches to three-dimensional photonic crystals. *Advanced Materials*. **13**: 409-413 (2009).
- A. Blanco, E. Chomski, S. Grabtchak, M. Ibisate, S. John, S.W. Leonard, C. Lopez, F. Meseguer, H. Miguez, J.P. Mondia, G.A. Ozin, O. Toader and H.M. van Driel, Large-scale synthesis of a silicon photonic crystal with a complete three-dimensional bandgap near 1.5 micrometres. *Nature*. 405: 437-440 (2000).
- 4. Y.A. Vlasov, X.-Z. Bo, J.C. Sturm and D.J. Norris, On-chip natural assembly of silicon photonic bandgap crystals. *Nature*. **414**: 289-293 (2001).
- 5. N. Huang, L.J. Martinez, E. Jaquay, A. Nakano and M.L. Povinelli, Optical epitaxial growth of gold nanoparticle arrays. *Nano Letters*. (2015).
- 6. F.X. Redl, K.S. Cho, C.B. Murray and S. O'Brien, Three-dimensional binary superlattices of magnetic nanocrystals and semiconductor quantum dots. *Nature*. **423**: 968-971 (2003).
- 7. A. van Blaaderen, R. Ruel and P. Wiltzius, Tempate-directed colloidal crystallization. *Nature*. **385**: 321-324 (1997).
- 8. J. Aizenberg, A.J. Black and G.M. Whitesides, Control of crystal nucleation by patterned selfassembled monolayers. *Nature*. **398**: 495-498 (1999).
- 9. Y. Yin, Y. Lu, B. Gates and Y. Xia, Template-assisted self-assembly: a practical route to complex aggregates of monodispersed colloids with well-defined sizes, shapes, and structures. *Journal of the American Chemical Society*. **123**: 8718-8729 (2001).

- 10. J.H. Lee, Q. Wu and W. Park, Metal nanocluster metamaterial fabricated by the colloidal selfassembly. *Opt. Lett.* **34**: 443-445 (2009).
- 11. C.A. Mejia, A. Dutt and M.L. Povinelli, Light-assisted templated self assembly using photonic crystal slabs. *Optics Express*. **19**: 11422-11428 (2011).
- 12. E. Jaquay, L.J. Martinez, C.A. Mejia and M.L. Povinelli, Light-assisted, templated self assembly using a photonic-crystal slab. *Nano Letters*. **13**: 2290-2294 (2013).
- 13. D. Erickson, Serey, X., Chen, Y.-F., Mandal, S., Nanomanipulation using near field photonics. *Lab Chip.* **11**: 995-1009 (2011).
- 14. E. Jaquay, L.J. Martinez, N. Huang, C.A. Mejia, D. Sarkar and M.L. Povinelli, Light-assisted, templated self assembly of gold nanoparticle chains. *Nano Letters*. **14**: 5184-5188 (2014).
- 15. J. Ma, L.J. Martinez and M.L. Povinelli, Optical trapping via guided resonance modes in a Slot-Suzuki-phase photonic crystal lattice. *Optics Express.* **20**: 6816-6824 (2012).
- 16. L.J. Martínez, N. Huang, J. Ma, C. Lin, E. Jaquay and M.L. Povinelli, Design and optical characterization of high-Q guided resonant modes in the slot-graphite photonic crystal lattice. *Optics Express* **21**: 30975 ((2013).).
- 17. A. Krishnan, N. Huang, S.-H. Wu, L.J. Martínez and M.L. Povinelli, Enhanced and selective optical trapping in a slot-graphite photonic crystal. *Optics Express*. **24**: 23271-23279 (2016).
- 18. R.D. Astumian, Thermodynamics and kinetics of a Brownian motor. *Science*. **276**: 917-922 (1997).
- 19. T.A.J. Duke and R.H. Austin, Microfabricated sieve for the continuous sorting of macromolecules. *Physical Review Letters*. **80**: 1552-1555 (1998).
- 20. L.R. Huang, J.O. Tegenfeldt, J.J. Kraeft, J.C. Sturm, R.H. Austin and E.C. Cox, A DNA prism for highspeed continuous fractionation of large DNA molecules. *Nature Biotechnology*. **20**: 1048-1051 (2002).
- 21. R.L. Smith, G.C. Spalding, K. Dholakia and M.P. MacDonald, Colloidal sorting in dynamic optical lattices. *Journal of Optics A: Pure and Applied Optics*. **9**: S134-S138 (2007).
- 22. S.-H. Wu, N. Huang, E. Jaquay and M.L. Povinelli, Near-field, on-chip optical Brownian ratchets. *Nano Letters*. **16**: 5261-5266 (2016).