REPORT DOCUMENTATION PAGE

Form Approved OMB NO. 0704-0188

1. REPORT DATE (DD-MM-YYYY)	2. REPORT TYPE	3. DATES COVERED (From - To)
22-02-2017	Final Report	15-Sep-2011 - 14-Jun-2015

4. TITLE AND SUBTITLE
Final Report: Frequency Agile Plasmonic Antennas and Sensors

5a. CONTRACT NUMBER
W911NF-11-1-0447

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER
611102

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

6. AUTHORS
Xiaoqin Li, Andrea Alu, Gennady Shvets

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES
University of Texas at Austin
101 East 27th Street
Suite 5.300
Austin, TX 78712 -1532

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES)
U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSOR/MONITOR'S ACRONYM(S)
ARO

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
60611-EL.7

12. DISTRIBUTION AVAILIBILITY STATEMENT
Approved for Public Release; Distribution Unlimited

13. SUPPLEMENTARY NOTES
The views, opinions and/or findings contained in this report are those of the author(s) and should not contrued as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT
As increasing demand to faster and smaller electronic and photonic devices, plasmonic technology which has promising approach to control light at length scale well below the optical diffraction limit has emerged. The small length scale of plamsmonic device, however, brings serious challenges in assembling, designing, and characterizing. The objective of this program is to develop an integrated method for assembly and characterization of individual nanostructures, to explore new design principles for plasmonic photonic devices, and to demonstrate prototypical devices to verify the effectiveness of both theoretical simulation and experimental approaches

15. SUBJECT TERMS
plasmonics, sensors, nanophotonics, experiments, theory

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			Xiaoqin Li
UU	UU	UU	UU		19b. TELEPHONE NUMBER 512-471-2063

Report Title

Final Report: Frequency Agile Plasmonic Antennas and Sensors

ABSTRACT

As increasing demand to faster and smaller electronic and photonic devices, plasmonic technology which has promising approach to control light at length scale well below the optical diffraction limit has emerged. The small length scale of plamsmonic device, however, brings serious challenges in assembling, designing, and characterizing. The objective of this program is to develop an integrated method for assembly and characterization of individual nanostructures, to explore new design principles for plasmonic photonic devices, and to demonstrate prototypical devices to verify the effectiveness of both theoretical simulation and experimental approaches.

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
02/22/2017	6	Thomas Hartsfield, Wei-Shun Chang, Seung-Cheol Yang, Tzuhsuan Ma, Jinwei Shi, Liuyang Sun, Gennady Shvets, Stephan Link, Xiaoqin Li. Single quantum dot controls a plasmonic cavity's scattering and anisotropy, Proceedings of the National Academy of Sciences, ( ): 12288. doi:
02/22/2017	5	Chun-Yuan Wang, Hung-Ying Chen, Liuyang Sun, Wei-Liang Chen, Yu-Ming Chang, Hyeyoung Ahn, Xiaoqin Li, Shangjr Gwo. Giant colloidal silver crystals for low-loss linear and nonlinear plasmonics, Nature Communications, ( ): 7734. 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

Number of Presentations: 0.00

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

Received Paper

TOTAL:

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

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

Received Paper

TOTAL:

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

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Book

TOTAL:

Received	Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

Xiaoqin Li and Andrea Alu have been elected as APS fellows

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Thomas, Harstfield	0.50	
Liuyang, Sun	0.10	
Chihhui Wu	0.10	
FTE Equivalent:	**0.70**	
Total Number:	**3**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Xiaoqin Li	0.08	
FTE Equivalent:	**0.08**	
Total Number:	**1**	

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: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

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:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

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:...... 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Thomas Hartsfield

Total Number: **1**

Names of other research staff

NAME	PERCENT_SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

see attachement

Technology Transfer

Final Report for ARO grant W911NF-11-1-0447

Frequency Agile Plasmonic Antennas and Sensors

PI: Xiaoqin (Elaine) Li
Physics department, the University of Texas-Austin, Austin, TX, 78712.
Tel: 512-471-2063; Fax: 512-471-1005; Email: elaineli@physics.utexas.edu
Co-PI: Gennady Shvets
Physics department, the University of Texas-Austin, Austin, TX, 78712. Email: gena@physics.utexas.edu
Co-PI: Andrea Alu
ECE department, the University of Texas-Austin, Austin, TX, 78712, Email: alu@mail.utexas.edu

Scientific Progress Report:

A. Objective

As increasing demand to faster and smaller electronic and photonic devices, plasmonic technology which has promising approach to control light at length scale well below the optical diffraction limit has emerged. The small length scale of plamsmonic device, however, brings serious challenges in assembling, designing, and characterizing. The objective of this proposal is to develop an integrated method for assembly and characterization of individual nanostructures, to explore new design principles for plasmonic photonic devices, and to demonstrate prototypical devices to verify the effectiveness of both theoretical simulation and experimental approaches.

B. The importance of the proposed research

We propose to use nanomanipulation method, which is based on based on atomic force microscopy (AFM), to assemble plasmonicnanophotonic device in a reconfigurable manner and to characterize these devices using optical dark-field scattering spectra. In addition to the development of the methodology, the expected outcome of the project includes the demonstration of two novel photonic devices. The first device is a **frequency agile nanoantenna**. Different from plasmonic waveguides, which have short propagation lengths limited by material loss, antennas may serve as an alternative approach to transfer energy/ optical signals in plasmonic circuits via free-space radiation. The second device is a **novel sensor based on optical dark modes** in nanorods. Dark modes can effectively enhance fields and store energy, therefore they may find applications in sensing as well as lasing and switching. The design of these novel plasmonic devices will take advantage of optical circuit element concepts and explore unique near field couplings between metallic nanostructures. We have accomplished essentially all the proposed experiments in the funding period.

Our research collaboration includes an experimental group led by the PI (Prof. Xiaoqin, Elaine, Li) and two theoretical groups led by co-PIs (Prof. Shvetsand Prof. Alu). The PI, Prof. Li, is an expert in nanophotonics. Her group has extensive experience in assembling and characterizing individual photonic nanostructures. Prof. Alu has pioneered extending the lumped circuit element concept to photonic devices, with particular interest in optical antennas. Prof. Shvets has made major contributions to important theoretical concepts such as Fano resonances in plasmonic nanoparticles (NPs) and metamaterials as well as highly asymmetric and bi-anisotropic plasmonic structures.

C. Accomplishment

In this final report, we focus on one accomplishment that represented the highlight of our program. Other important accomplishments have been summarized in previous annual report. We experimentally demonstrate for the first time that a single semiconductor quantum dot placed in nanometer-scale proximity of a plasmonic cavity can be used to control the scattering spectrum and anisotropy of the latter. This work was published in Proceedings of the National Academy of Sciences in 2015.

Many quantum network and information processing schemes require the enhanced light-matter interaction between a single quantum emitter and a cavity, enabling the effective conversion between photonic and matter-based quantum states. Those cavity-quantum electrodynamics (QED) effects require a high Purcell factor $F_P \propto Q/V$, where Q is the quality factor, and V is the volume of the cavity mode.

Prior experiments exploring cavity QED effects associated with single emitters coupled to plasmonic cavities or waveguides focused almost exclusively on the observations of reducing the emitter's lifetimes. The possibility of controlling the scattering of a plasmonic nanocavity by a single (and inherently quantum and nonlinear) two-level system has also been proposed but never experimentally observed.

The strongly coupled MNP-QD hybrid structure is assembled into a well-controlled geometry using the technique of AFM nanomanipulation. The strong coupling between the MNP and QD is experimentally confirmed by measuring the exciton lifetime. Analyzing the polarization and spectral properties of light scattered by the MNP-QD hybrid, we observe that the overall plasmonic cavity scattering is significantly modified over a broad spectral range. A Fano resonance spectrally aligned with the QD's quantized exciton resonance is clearly identified when the polarization of the scattered photon is along the Fano axis connecting the MNP's center with the QD. The anisotropic scattering spectrum observed in our experiments suggests that a polarization-controlled, versatile quantum light source may be realized in this simple QD-MNP cavity system.

The calculated polarization-resolved scattering spectra by the QD-MNP (diameters: $2r_{QD} = 6$nm and $2r_{MNP} = 30$nm) hybrid are shown in Fig.1(a) for three polarization angles ϕ_A of the analyzer placed in the collection path of the scattering signal to mimic the experimental setup. In the absence of the QD, all scattering spectra from a single MNP are independent of ϕ_A and possess a single broad peak at $\lambda_{MNP} \approx 520$nm corresponding to the plasmonic dipole resonance of the MNP. The introduction of a QD under the MNP, with the separation gap of $g = 1nm$, modifies the scattering spectrum: a sharp Fano feature emerges at the exciton transition wavelength $\lambda_{QD} = 550$nm.

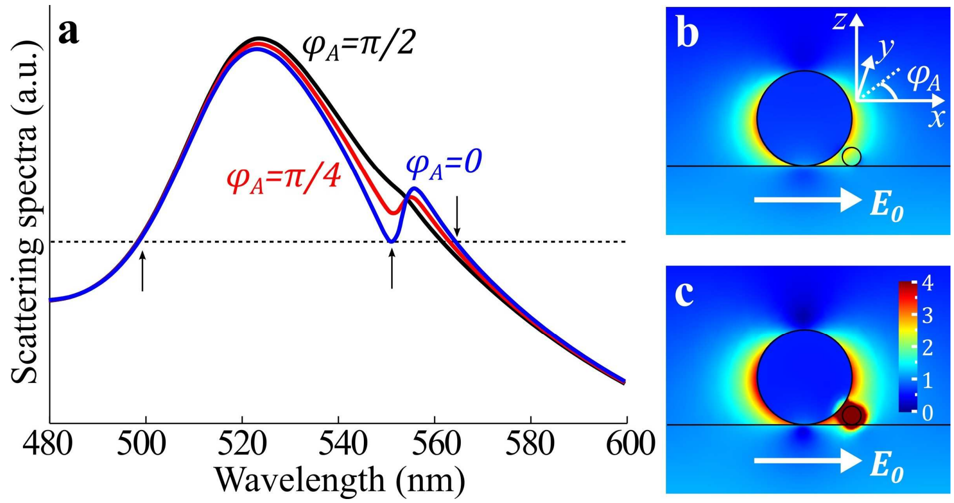

Fig 1: **Calculation demonstrating how the near-field coupling modifies the far-field scattering spectra of a NP-QD hybrid structure placed on a glass substrate.**(a) The scattering spectra of a NP-QD hybrid excited by the un-polarized evanescent wave coming from the glass substrate side in all azimuthal angles. The angle φ_A indicates the orientation of the analyzer in the path of the scattered light. Fano feature is the most (least) prominent when the orientation of the analyzer is parallel (perpendicular) with the in-plane component of the Fano axis, which connects the QD and MNP centers. (b) and (c) shows the field near the NP-QD hybrid at 500 nm and 552 nm, respectively, as indicated by the black arrows in the scattering spectrum at $\varphi_A = 0$ in (a). The scattering signal at these two wavelengths is the same (indicated by the dotted line on the blue curve) while the MNP is excited much more strongly in Fig 1c. This wavelength dependence proves that the presence of the QD indeed controls the MNP's scattering and anisotropy resonantly.

The ability of a single QD to modify the scattering spectrum of a much larger MNP possessing a scattering cross section of $\left(r_{\mathrm{MNP}}/r_{\mathrm{QD}}\right)^6 \approx 15{,}000$-fold greater in magnitude than that of the QD seems surprising. Naively, one may expect the effect of a QD on the scattering spectrum of the hybrid system to be very small. As pointed out by a number of theoretical studies and our own numerical calculation shown in Fig. 9, this expectation is not correct.

To illustrate this point, the near-field distributions were calculated for $\lambda_1 = 500$ nm (Fig. 1b) and $\lambda_2 = 552$ nm (Fig. 1c), respectively. These two wavelengths were chosen because the scattering intensities are the same. The much higher (by almost a factor of 2) electric field induced on the MNP's surface at $\lambda = \lambda_2$, is offset by strong near-field depolarization (light-color area near the QD) of the MNP by the exciton's dipole. Because the electric field of a dipole rapidly decays with distance, such extreme de-polarization (which can be alternatively interpreted as the excitation of high-order multipoles of the MNP by an exciton) can only occur if the QD is placed within nanometers from the MNP.

We assemble the hybrid structure using the technique of AFM nanomanipulation. We first disperse MNPs and QDs on a glass substrate randomly. We then simultaneously obtain an AFM topography image and a photoluminescence (PL) image by scanning the sample on a home-built integrated AFM-confocal microscope. We locate isolated MNPs and QDs in close proximity via the AFM topography image and push a near-by MNP to approach the chosen QD. We then measure the lifetime to confirm that the MNP is indeed in the close proximity of the QD.

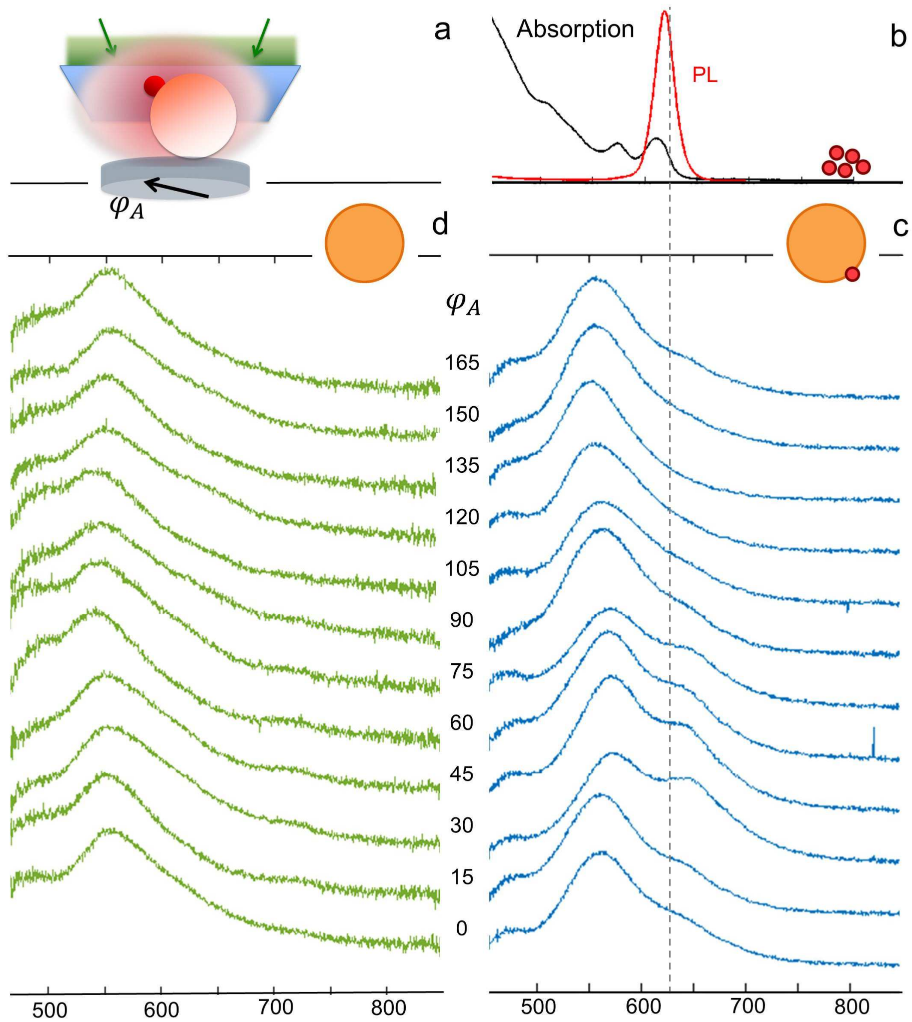

Fig. 2: The scattering and anisotropy of a plasmonic cavity (i.e. a MNP) controlled by a single QD. (a) Experimental schematics for scattering measurements; (b) Experimentally measured absorption (black) and PL emission (red) spectra from an ensemble of colloidal CdSe/ZnS core-shell QDs. (c) Measured scattering spectra of the assembled QD-MNP hybrid structure at different analyzer angles. The Fano resonance indicated by the dotted vertical line spectrally aligns with the lowest exciton state measured in the absorption from an ensemble of QDs. The Fano resonance is most pronounced at $\phi_A = 30°$. (d) Measured scattering spectra of a bare 30 nm diameter MNP at different analyzer angles. The small changes near 550 nm or below are likely due to deviations from a perfectly spherical shape of the MNP.

Measured ensemble absorption and PL emission spectra (taken in solution) of QDs are shown in Fig. 2b. The absorption spectrum features multiple discrete exciton resonances at lower energies and a continuous absorption spectrum at energies above the band gap of the crystal.

While all absorption resonances may influence the scattering spectrum of the hybrid structure, we focus on the lowest-energy exciton state centered near $\lambda_{QD} \approx 615$ nm with an ensemble-averaged spectral full-width at half-maximum (FWHM) $\Delta\lambda_{1/2}{\sim}15$ nm.

The dark-field scattering experiments are performed using a home-built optical system optimized for small MNP measurements (Fig. 2a). An un-polarized white light source incident in a conical geometry generates evanescent fields and excites the hybrid structure from all directions. An analyzer is placed in the scattered light's path to select the polarization of the collected scattering. A series of such spectra are displayed in Fig. 2c as a function of the analyzer angle. A very sharp Fano feature can be clearly observed for $\phi_A = \phi_1 = 30°$. Weaker Fano features are observed for other polarizer orientations, all in qualitative agreement with our theoretical predictions in Fig. 1a.

To further confirm that the polarization dependence in the scattering spectra of the hybrid structure indeed originates from the coupling between the QD and MNP, we show the scattering spectra of a bare MNP in Fig. 2d. The spectra do not display any Fano features or analyzer angle dependence in the spectral proximity of the exciton resonance λ_{QD}. Therefore, it is indeed the coupling between a single QD and the MNP that turns an otherwise isotropic plasmonic cavity into a strongly anisotropic one. Because a single quantum absorber achieves this effect, one can envision the proposed hybrid system as an experimental platform for observing a plasmonic cavity anisotropy controlled by optical nonlinearity at the single-photon level. The small angular variations of $S(\lambda, \phi_A)$ at shorter wavelengths (around 550nm or below) in Fig. 2c and Fig. 2d most likely arise from a small intrinsic deviation of the MNP's shape from an ideal sphere. Unlike the extrinsic anisotropy induced by the QD, it cannot be optically controlled and is not of interest for nonlinear quantum optics.

D. Conference Presentation:

1.F. Monticone, X. Li, and A. Alù, “Strong Optical Magnetism and Fano Resonances in Asymmetric PlasmonicMetamolecules,” 2013 IEEE International Symposium on Antennas and Propagation, Lake Buena Vista, FL, July 7-12, 2013.

2.N. MohammadiEstakhri, and A. Alù, “Controlling the Optical Wave Propagation Using Engineered NanoscaleMetasurfaces,” 2013 IEEE International Symposium on Antennas and Propagation, Lake Buena Vista, FL, July 7-12, 2013

3. P. Y. Chen, and A. Alù, “Nanoantenna-Enhanced Optical Heterodyning and Photoemission Nanodevices,” 2013 IEEE International Symposium on Antennas and Propagation, Lake Buena Vista, FL, July 7-12, 2013

4. N. MohammadiEstakhri, and A. Alù, “Graded Metareflectors for Wave Manipulation and Control at the Nanoscale,” URSI National Radio Science Meeting, Boulder, CO, January 8-11, 2014.

5. Y. Wu, C. Zhang, J. Kim, M. Zhang, C. K. Shih, X. Li, Y. Zhao, N. MohammadiEstakhri, X. X. Liu, A. Alù, and G. Pribil, “Extraordinary SPP Propagation Distance in Epitaxially Grown Silver Film,” 2014 APS March Meeting, Denver, CO, March 3-7, 2014.

6. F. Shafiei, F. Monticone, K. Q. Le, X. X. Liu, T. Hartsfield, A. Alù, and X. Li, “Plasmonic Magnetic Nanostructure,” 2014 APS March Meeting, Denver, CO, March 3-7, 2014.

7. J. Shi, S. Elias, F. Monticone, Y. Wu, D. Ratchford, X. Li, and A. Alù, “Assembling Three-Dimensional Optical Stereo-Nanocircuits,” 2014 APS March Meeting, Denver, CO, March 3-7, 2014.

8. N. MoahhamdiEstakhri, and A. Alù, “Towards Efficient Nanoscale Wave Manipulation with Graded PlasmonicMetareflectors,” 2014 MRS Spring Meeting, San Francisco, CA, April 21-25, 2014.

E. Publication in peer-reviewed journals

1. J. Shi*, S. Elias*, F. Monticone*, Y. Wu, D. Ratchford, X. Li, and A. Alù, “Modular Assembly of Optical Nanocircuits,” Nature Communications, Vol. 5, No. 3896 (8 pages), May 29, 2014.
2. Y. Zhao, J. Shi, L. Sun, X. Li, and A. Alù, “Alignment-Free Three-Dimensional Optical Metamaterials,” Advanced Materials, Vol. 26, No. 9, pp. 1439-1445, 2014.
3. N. MohammadiEstakhri, and A. Alù, “Manipulating Optical Reflections Using Engineered NanoscaleMetasurfaces,” Physical Review B, Vol. 89, No. 23, 235419 (8 pages), June 16, 2014.
4. P. Y. Chen, and A. Alù, “A Terahertz Photomixer Based on PlasmonicNanoantennas Coupled to a Graphene Emitter,” Nanotechnology, Vol. 24, No. 45, 455202, 2013.
5. “Giant Colloidal Silver Crystals for Low-Loss Linear and Nonlinear Plasmonics”, Chun-Yuan Wang, Hung-Ying Chen, Liuyang Sun, Wei-Liang Chen, Yu-Ming Chang, Hyeyoung Ahn, Xiaoqin Li and Shangjr Gwo, Nature Communications, 6, 7734, 2015.
6. “Single Quantum Dot Controls a Plasmonic Cavity’s Scattering and Anisotropy” Thomas Hartsfield, Wei-Shun Chang, Seung-Cheol Yang, Tzuhsuan Ma, Jinwei Shi, Liuyang Sun, Gennady Shvets, Stephan Link, and Xiaoqin Li, PNAS 112, 12288, 2015.