Nanoplasmonics for Ultrafast Coherent Control of Optical Fields

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Report Date: May 2014

Final Report from 1 September 2012 to 28 February 2014

REPORT DOCUMENTATION PAGE

Form Approved OMB No. 0704-0188

1. REPORT DATE (DD-MM-YYYY)	2. REPORT TYPE	3. DATES COVERED (From – To)
1 May 2014	Final Report	1 September 2012 –28 February 2014

4. TITLE AND SUBTITLE	5a. CONTRACT NUMBER
Nanoplasmonics for Ultrafast Coherent Control of Optical Fields	**FA8655-12-1-2091**

5b. GRANT NUMBER

Grant 12-2091

5c. PROGRAM ELEMENT NUMBER

61102F

6. AUTHOR(S)

Khalid Rajab

5d. PROJECT NUMBER

5d. TASK NUMBER

5e. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
QUEEN MARY & WESTFIELD COLLEGE
UNIVERSITY OF LONDON
MILE END ROAD
LONDON, E1 4NS UNITED KINGDOM

8. PERFORMING ORGANIZATION REPORT NUMBER

N/A

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

EOARD
Unit 4515
APO AE 09421-4515

10. SPONSOR/MONITOR'S ACRONYM(S)

AFRL/AFOSR/IOE (EOARD)

11. SPONSOR/MONITOR'S REPORT NUMBER(S)

AFRL-AFOSR-UK-TR-2014-0031

12. DISTRIBUTION/AVAILABILITY STATEMENT

Distribution A: Approved for public release; distribution is unlimited.

13. SUPPLEMENTARY NOTES

14. ABSTRACT

This one-year project investigated plasmonic nanoantennas, comparing snowflake and log-periodic designs, and applied developed tools to nanoantenna arrays, "chirped signals", and nanoantenna responses with dielectric and graphene loads. The project was seeking to maximize bandwidth and reduce temporal pulse width. Some findings: time-domain methods enabled modeling of nonlinearity and chirp, a circularly polarized pulse train can sequentially excite arms of an antenna to create a center-point pulse train, closely-spaced antennas can be resolved, log-periodic antennas have increased bandwidth vs snowflake, and the high surface impedance inherent to graphene results in high field concentrations.

15. SUBJECT TERMS

EOARD, nanoantennas, nanoantenna arrays, chirped signals, log-periodic, graphene

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18, NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			Victor Putz
UNCLAS	UNCLAS	UNCLAS	SAR	7	19b. TELEPHONE NUMBER (Include area code) +44 (0)1895 616013

Standard Form 298 (Rev. 8/98)
Prescribed by ANSI Std. Z39-18

Final Report for EOARD/AFRL Project Grant FA8655-12-1-2091

"NANOPLASMONICS FOR ULTRAFAST COHERENT CONTROL OF OPTICAL FIELDS"

Final work summary, May 2014

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Background

The project funded an investigation of one year with the following main objectives:

- Develop time-domain modelling tools for investigation of plasmonic nanoantennas, and comparison of snowflake and log-periodic nanoantennas.
- Application of developed tools to nanoantenna arrays.
- Stimulation of nanoantennas with temporally- and polarization-controlled waveforms for field manipulation and control.
- Nanoantenna response modification using dielectric/graphene loads.

With ultrafast temporal resolutions on the order of attoseconds, nanoplasmonics are potentially the fastest in optics. Nanoplasmonics involves the investigation of surface plasmons (SPs), which are excited by the interaction of electromagnetic waves with metallic nanostructures. Plasmonic nanoantennas, made of metals such as gold, have been proposed as a means of controlling and manipulating femtosecond pulses. In this project we have investigated nanoantenna designs, to maximise bandwidth and thus reduce temporal pulse widths. We have also examined their ability to manipulate and control light, subject to the shaping of incident wave polarization and temporal responses via pulse chirping. Techniques developed have been applied to single pulses and pulse trains. We summarise the results as follows: application of full-wave time-domain modelling tools; incident wave polarization and waveform shaping; comparison of snowflake and log-periodic nanoantennas; effect of dielectric or graphene loads.

Full-wave time-domain modelling

The structures were modelled using in-house finite-difference time domain codes, and compared to CST Microwave Studio (time domain) and Ansoft HFSS (frequency domain) simulations. In all cases the results matched reasonably well, on the condition that a sufficiently fine mesh was selected. **Care must be taken in handling spatial boundaries involving dispersive media.** The nanoantennas were composed of gold, which exhibits dispersive, plasmonic behavior at the frequencies of interest. This could be approximated with a Drude model, and can be incorporated into the FDTD code using well known techniques. **The strength of the time-domain methods lies in their ability to model phenomena such as nonlinearity and chirp quite naturally.** We have also used the full-wave simulations to enable investigation of the field distributions and concentrations (Figure 1).
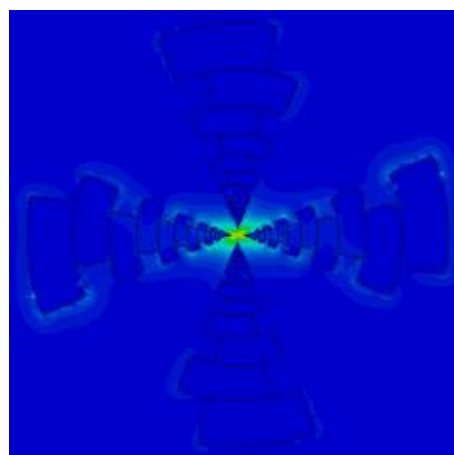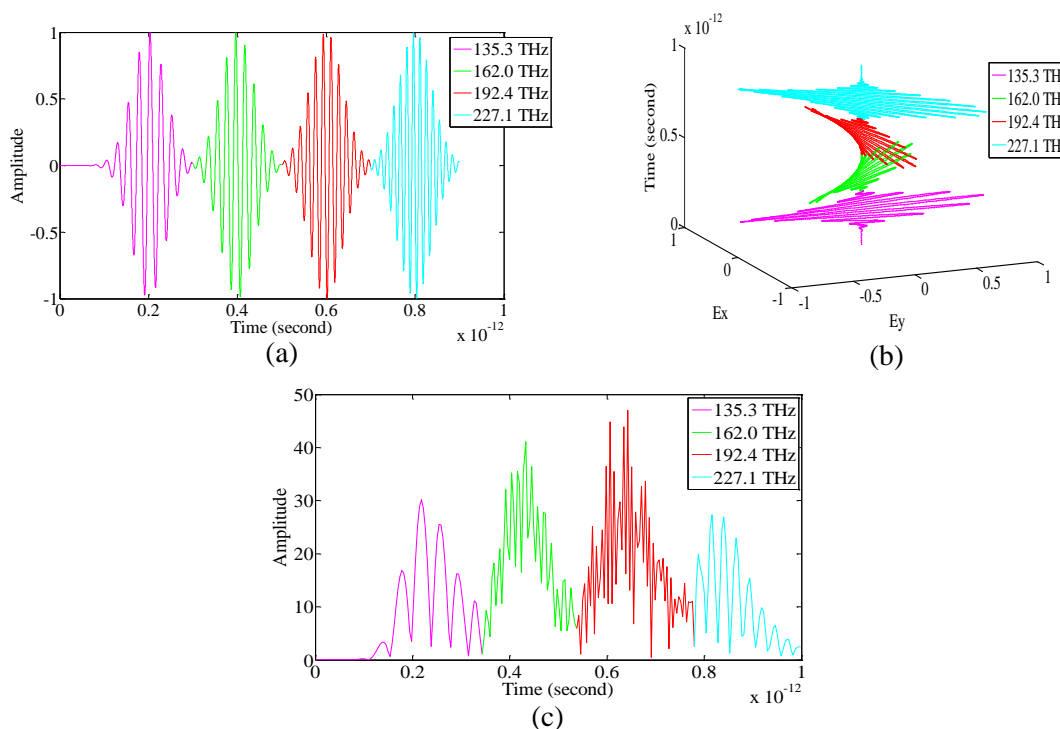

Figure 1 - Field distribution of a log-periodic nanoantenna

Nanostructure waveform synthesizing

Waveform synthesis for nanostructure excitation is engineered based on a priori knowledge of their respective resonant frequencies. For analysis of a structure such as the snowflake nanoantenna, a wide band Gaussian pulse

acts as an exciting waveform to investigate the resonant frequencies of the dipole elements, either individually or in a cluster. With this spectral information determined, a circularly polarized pulse train is then synthesized based on the resonant frequencies to control excitation of the field distribution and concentration. A multi-band energy concentration is realized via such a configuration.

Circularly polarized waveforms exhibit polarizations that change with time, and so with a nanoantenna that is responsive to different polarizations one may take advantage of this to improve control of the field concentrations. A snowflake nanoantenna for example, may have dipolar arms of different lengths that are responsive to different frequencies and polarizations. **A circularly polarized pulse train can excite the different arms in sequence to create a pulse train at the center-point of the nanoantenna** (Figure 2). It is important to note that the field concentrations are dependent not just on the structural geometry, but also on the waveform of the excitation.

Figure 2 – (a) Gaussian sine pulse train with corresponding frequencies. (b) Circularly polarized waveform. (c) Resultant pulse train at center of nanoantenna.

Concentration of energy with a spatial resolution on the nanoscale and at the temporal resolution on the order of femtoseconds or even attoseconds are critically important in the applications of spectroscopy, quantum system control, controlled chemical reaction, multiplexing and logic switching. Due to the size limitation, conventional techniques fail in such applications. They either do not fit in the nanostructure system or perturb the electromagnetic field distribution when the probes are in the near field of the nano devices. An elegant solution to this problem is to manipulate the illumination pulse transmitted in the far field of the subject. The positive chirp, negative chirp and Gaussian pulse waveforms are used as the excitations. A temporal resolution of 24 fs, which is of the order of the typical semiconductor switching speed, and a spatial resolution (dipole element spacing 50nm) of $\lambda/40 \sim \lambda/24$, which is not far from the size of typical organic molecules (of the order of 1 nm [32]), are achieved.

Linear chirp, where the frequency of the waveform increases (or decreases) linearly with time, can quite easily be synthesized using time-domain techniques. Despite the fact that the overall frequency profile will be quite similar to that of a Gaussian waveform, **we have shown that with careful design we can resolve dipole nanoantennas that are closely spaced, with spatial resolutions of $\lambda/40 \sim \lambda/24$**. For comparison, organic molecules have size typically on the order of around 1 nm. Furthermore, temporal resolutions of 24 fs – on the order of the typical semiconductor switching speed – are also obtained. To demonstrate the resolving

capabilities of this waveform it is compared to a Gaussian pulse with identical frequency profile, in which the features are not successfully resolved (Figure 3).
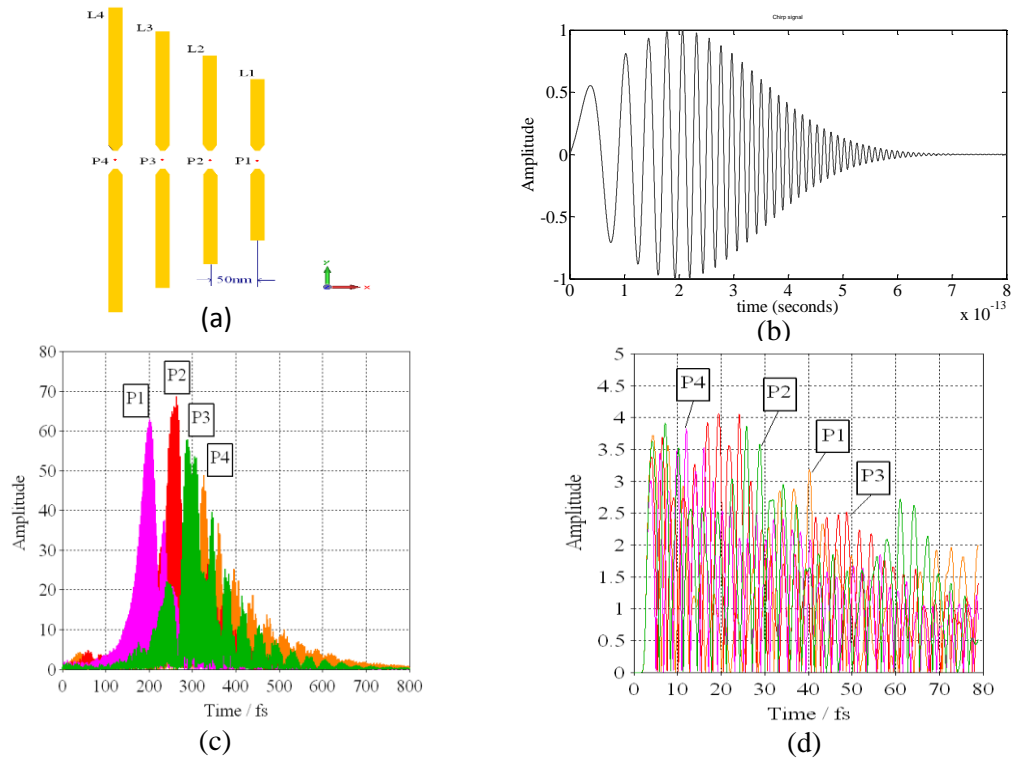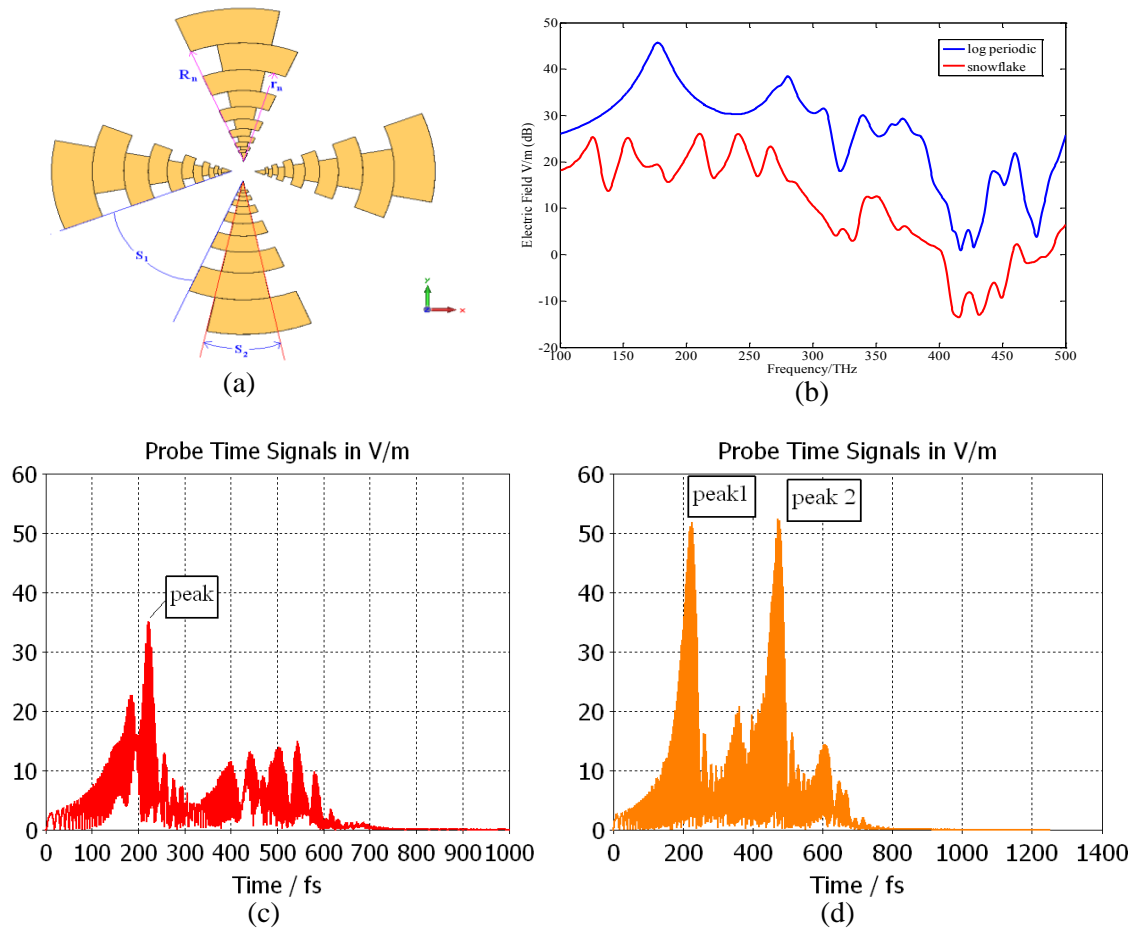

(a)

(b)

(c)

(d)

Figure 3 - (a) Linear array of dipoles. (b) Positive linear chirp excitation signal. (c) Field at center-gap due to positive linear chirp excitation. (c) Field at center-gap due to unchirped Gaussian excitation.

Comparison of snowflake and log-periodic nanoantennas

The development of log-periodic nanoantennas has been considered for comparison to snowflakes. Originally designed for broadband operation at microwave frequencies, their geometrical structures give them very wideband performance. We have simulated these structures in detail, and demonstrated that **the increased inherent bandwidth results in magnification of the field concentration in comparison to the snowflake antennas**. This improvement can be over 10dB over a range of frequencies (Figure 4).
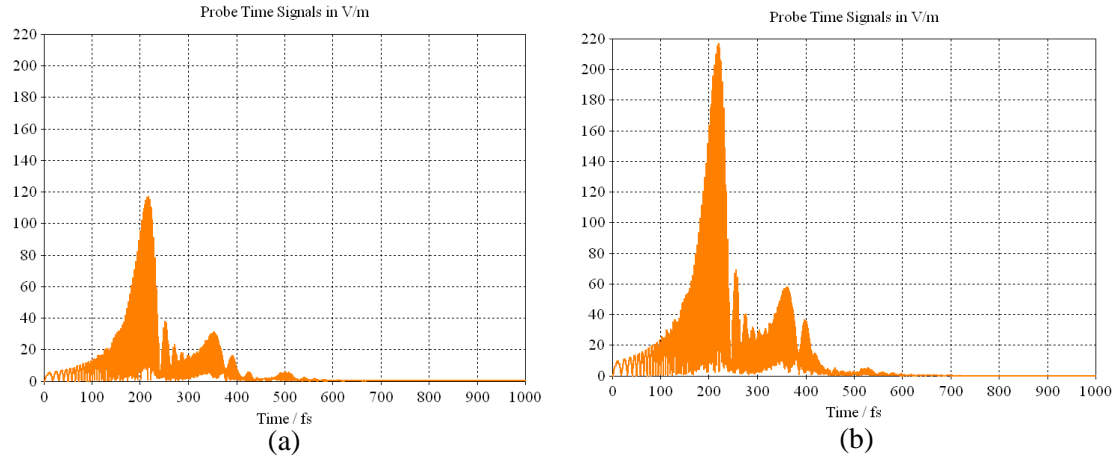
Figure 4 - (a) Log-periodic nanoantenna geometry. (b) Comparison of log-periodic to snowflake nanoantenna. (c) Pulse train detection using snowflake. (d) Pulse train detection using log-periodic nanoantenna.

Effects of graphene and dielectric loads

Considering the nanostructure as a *nanocircuit*, there is an intrinsic impedance associated with it. The nanocircuit is terminated with a load at the center (i.e. the gap is filled with a load), and so a field intensity change at the gap centre is observed. This allows further modification of the field concentration, based on the nature of the loading material.

The effects of various nanoantenna loads has been investigated, including high permittivity dielectric discs and graphene monolayers. For this investigation, graphene was implemented using a surface impedance model which was incorporated into the simulations. Although the dielectric loads led to relatively little change in the resonant profile, **the high surface impedance inherent to graphene results in high field concentrations** (Figure 5). Based on the nanocircuit model, graphene shows the best impedance match to the nanoantenna. We believe that this is an interesting avenue of research that warrants further investigation.

The above stated areas of research act as bridges toward many more application level investigations, such as on-chip and intra-chip wireless nano-links, light ray conversion, nano-sensing, nanoscale spectroscopy, quantum communication, and wireless optical power transmission.

Figure 5 - Time signals at the gap center of the log periodic antenna using chirp excitation. Two loading scenarios: (a) air gap; (b) high surface impedance material

Dissemination of results

A manuscript (attached) is currently undergoing final edits, for submission for publication in a leading journal. We are currently considering targeting *Physical Review A*.