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Advanced Metacrystal Media for Aerospace applications

David Smith
DUKE UNIVERSITY

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Final Report

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14. ABSTRACT We propose a broad and comprehensive program to investigate a variety of topics relating to advanced metamaterials. We have selected topics across the electromagnetic spectrum—with a particular emphasis in the microwave regime—in which we will apply a variety of design, fabrication and characterization methods to develop new metamaterial paradigms. In one research thrust, we propose the development of hybrid, anisotropic MetaCrystals for optical and quasi-optical devices. MetaCrystals are the metamaterial analog of natural crystals that serve as critical components in optical technologie					
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Advanced MetaCrystal Media for Aerospace Applications

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PI: David R. Smith

FINAL REPORT

Final Report

The purpose of the MetaCrystal Media program was to develop and investigate hybrid metamaterials—metamaterials that could provide advanced functionality by incorporating core materials with properties of interest—such as nonlinearity or tunability—into artificially structured metamaterials. Metamaterials provide a means of creating unique electromagnetic response, which can be used to manipulate and control electromagnetic waves propagation. In addition, metamaterials can concentrate electromagnetic or optical fields into highly localized regions where they can interact strongly with materials. This interaction can be harnessed to create multifunctional metamaterials.

Nonlinear Metamaterials

One of the first studies performed within this program was the use of nonlinear materials to form artificial nonlinear optical materials. We developed a comprehensive analytic theory and design methodology for nonlinear metamaterials, as well as a suite of computational techniques that allowed for the precise design and characterization of nonlinear metamaterial composites. We applied these tools in the design and demonstration of numerous nonlinear metamaterial structures, including the first experimental demonstration of phase matching with a negative index nonlinear metamaterial and the demonstration of harmonic generation in a material with index near zero. The initial metamaterial samples were made to operate at microwave frequencies, and utilized varactor diodes as the nonlinear component. By placing varactor diodes across the capacitive gaps in metamaterial samples, an artificial nonlinear metacrystal can be formed and its properties compared with theoretical predictions. To support this effort, a retrieval method was devised, allowing the assignment of the effective nonlinear susceptibility to a metamaterial sample based on the harmonics produced when excited by a fundamental wave. Using this retrieval, near exact agreement was demonstrated between theory and experimentally measured samples. The results of this study proved the ability to design and characterize artificial nonlinear crystals.

Nonlinear Optical Plasmonic Metamaterials

After demonstrating the basic concepts behind nonlinear metamaterial design, we began a program to develop an understanding of nonlinear metamaterials at infrared (IR) and visible wavelengths. At these wavelengths, metal response becomes significantly more complicated, and more detailed models of electron response must be incorporated into full-wave simulations. Moreover, metals possess some of the largest third-order nonlinear susceptibilities in materials, and can also yield very large second-order nonlinear susceptibilities when structured on the nanoscale (due to surface effects). As part of our program, we developed a set of numerical tools to investigate the nonlinear response of optical metamaterials, treating the metal using a semiclassical model of electron response. In particular, a hydrodynamic model was applied, in which the electron gas is treated as non-compressible and an electron pressure term is added to



the equation of motion. Within this model, nonlinear terms within the hydrodynamic equations can be retained and used to predict various nonlinear effects. We applied the model to understand the origin of second harmonic generation in metamaterial structures at optical wavelengths, confirming the empirically obtained experimental results of the Karlsruhe group (M. Wegener). A variety of nonlinear metallic configurations were considered as part of this effort, which continues now in the follow-on AFOSR program.

Nonlocal Response and the Extreme Coupling Limit

In the quest to understand the inherent nonlinear response of metals at optical wavelengths, we found it was necessary to make use of the hydrodynamic model of electron response. This model introduces an electron pressure term into the equations that results in the effective dielectric function being non-local—that is, having a dependence on wave-vector in addition to frequency. The nonlocal response of the dielectric function requires the imposition of additional boundary conditions at the interface between the nonlocal dielectric and other materials, resulting in a more complicated computational problem. As part of this program, we developed a simulation approach for nonlocal media, and made use of the approach to compute the properties of nonlinear composites as described in the previous section. However, the nonlocal electron response also has implications for linear optical scattering, though those effects are typically insignificant.

To probe the nonlocal response directly, we made use of the film-coupled nanoparticle system, using self-assembled monolayers to create uniform spacers between a collection of nanoparticles and a gold film. The layer-by-layer assembly technique allowed control over the spacing in increments of roughly a few Ångströms. Even greater control could be achieved using carbon chains terminated with amine groups on one side and thiol groups on the other, such that spacer layers could be produced with control to the Ångström level. Studying the plasmon resonance shift associated with this film-coupled nanoparticle system as a function of spacer layer, we were able to probe the extreme coupling limit, where the nanoparticles were spaced at distances between 0.5 and ~25 nm, with Ångström level precision. Approach curves indicated a failure of the classical model of electron response, and consistency with the nonlocal response (contained in the hydrodynamic model). While there is much to be done to further understand this important phenomena, the experiment (published in *Science*) brought to the forefront the need for new models for the metal response in nanostructured plasmonic systems having extreme feature sizes (e.g., sub-nanometer gaps).

Film-Coupled NanoCube System

The film-coupled nanoparticle system provides an unprecedented, unparalleled means of controlling the enhancement region in plasmonics and optical metamaterials. The entire range of plasmonic effects, including surface enhanced Raman scattering (SERS), fluorescence enhancement, nonlinear enhancement and many others—all such effects are tied to the details of the field enhancement that occurs in the sub-nanometer gaps between plasmonic nanoparticles. Because the film-coupled nanoparticle system relies on planar fabrication for the gap (or spacer) layers, a variety of surface chemistries can be used to create films with precisely controlled thickness; alternatively, deposition techniques, such as atomic layer deposition (ALD) can be leveraged to achieve Ångström level control over the layer thicknesses. In this program, we have investigated both techniques and shown that control can be exerted over the enhancement region to the sub-nanometer scale.



Film-coupled nanospheres were used to demonstrate the impact of non-local response; however, the film-coupled nanosphere system does not provide many degrees of freedom to enhance other phenomena. For example, the field enhancement is tied directly to the plasmon resonance shift in the film-coupled nanosphere system: If the sphere diameter is changed, or the gap, both the resonance and the field enhancement are changed, making it difficult to perform optimization where both the resonance and field enhancement need to be tuned separately.

The film-coupled nanocube system possesses entirely distinct properties from that of the film-coupled nanosphere. The nanocube, being a planar particle, supports transmission-line modes between the nanocube and film, whose resonance frequencies depend entirely on the nanocube width. The field enhancement is separately controlled via the gap thickness, allowing for enhancement effects to be tuned and optimized. As part of this program, we have developed a comprehensive theoretical framework for the optical patch antenna (of which the film-coupled nanocube is an example), as well as extending the simulation tools to accommodate this geometry. We have also performed an experiment showing the potential use of the nanocubes as a “perfect absorber” medium, with the advantage that such a medium can be achieved through self-assembly rather than lithography (this work was published in *Nature*). The film-coupled nanocube system can be used as the basis for ultra-bright light emitting diodes (LEDs) or efficient detectors. We have also performed a number of experimental studies to explore and confirm the unique modes associated with film-coupled nanocubes. This work continues on into the follow-on AFOSR program.

Metamaterial Computational Imaging System

As part of this program, we have investigated the use of metamaterials as the basis of a new type of aperture for computational imaging schemes. The metamaterial aperture consists of either a one dimensional waveguide (such as a microstrip) or a two dimensional waveguide (such as a parallel plate waveguide), with complementary metamaterial elements patterned into the upper conductor. Because these metamaterial elements resemble irises, they allow radiation to leak from the waveguide mode into the free space region to be imaged. The configuration is reminiscent of a leaky-wave antenna, however the sub-wavelength metamaterial elements are densely packed, and their resonances dominate the dispersive properties of the aperture. The metamaterial elements are patterned such that the resonance frequency of each of the elements is assigned randomly over the operating bandwidth (in this case, 18-26 GHz). As a function of frequency, then, the radiated mode pattern has low gain with a random set of nodes that move around in space as the frequency is varied. Because there is sufficient diversity in the modes, they can be used to encode distinct scene information; that is, an image can be formed simply by conducting a frequency sweep over a suitably large bandwidth, with the image reconstructed using standard computational image estimation.

The metamaterial imager concept has grown out of the legacy of AFOSR funding, starting with the complementary metamaterial waveguide structures initiated in a previous AFOSR program and expanded in the present program. The metamaterial aperture has potential advantages over conventional technology, in that it utilizes a small number of transceivers (possibly just one), and has no moving or scanning mechanical components. To confirm the imaging concept, we performed an imaging experiment using a one-dimensional aperture that could provide a low-resolution image in the range and one angular variable (1+1 dimensions). It was shown that a retroreflector could be tracked, at 10 frames-per-second, using just the frequency sweep (frequency diversity) and compressive imaging techniques. That is, the scene was vastly



undersampled for the experiment. This work was published in Science, and represents the first report of the frequency-diverse metamaterial imager.

Subsequently, a two-dimensional metamaterial imager system was built, and was used to demonstrate the ability to resolve full three-dimensional images (range+two angular variables). This system consisted of two metamaterial panels and a single low gain probe. Simple metal objects were imaged, showing that the metamaterial imager should be capable of ultimately state-of-the-art resolution.

During the time when this research was being conducted, representatives from the Department of Homeland Security and the Transportation Safety Division visited our lab and became interested in the imaging concept. This led to a very large program, jointly funded by DHS and TSA, to develop next-generation mm-wave imagers for aviation security. As an outgrowth of the AFOSR research, we are now deeply involved in the full system design of a functioning scanner, appropriate for current airport security requirements. As part of this program, we are building a full-scale demonstration unit (1.6 m x 1.6 m), with complete software support, standalone radio, and integrated automated threat detection (ATD) software. This work has also led to a spinoff company, Evolv Technology (Boston, MA), working to develop commercial units based on the underlying technology.

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