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# Metamaterials at the University of Southampton and beyond

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## Abstract

At the University of Southampton, research on metamaterials started in 2000 with a paper describing a metallic microstructure, comprising an ensemble of fully metallic 'molecules'. In the years since then, metamaterials have evolved from an approach for designing unusual electromagnetic properties by structuring matter at the sub-wavelength scale to become a universal paradigm for active functional media with optical properties on demand at any given point in time and space. Metamaterials are now recognized as a promising enabling technology and one of the most buoyant and exciting research disciplines at the crossroads between photonics and nanoscience. Many 'made in Southampton' concepts have influenced the global research community, and we are now working in collaboration with our sister research centre in Nanyang Technological University, Singapore. In this article, I provide a historical overview of the metamaterials research field, from early studies to recent developments through the lens of the Southampton group, and discuss promising future directions.

Keywords: metamaterials, nanophotonics, University of Southampton, Nanyang Technological University

(Some figures may appear in colour only in the online journal)

## Introduction

The metamaterial paradigm, without using this now fashionable term, was articulated by Richard Feynman in 1959, stating 'Up to now, we have been content to dig in the ground to find minerals ... What would happen if we could arrange the atoms one by one the way we want them? ... when we have some control of the arrangement of things on a small scale we will get an enormously greater range of possible properties that substances can have, and of different things that we can do' [1]. In essence, metamaterials are all about new functionalities that can be achieved with conventional materials by arranging them on a small scale. It has proved to be an extremely powerful concept.

Indeed, metamaterial studies have become one of the most fertile areas of science. What are metamaterials? Today this term

refers to artificial materials with electromagnetic properties on demand [2, 3]. Electromagnetic metamaterials can not only be designed to give certain *static* characteristics to the electromagnetic space, but now work is being done on developing *dynamic* metamaterials that can provide desired electromagnetic characteristics at a given point of time and space. Why are metamaterials so interesting? They can be fabricated top down, in particular using the tools and processes of nanotechnology, they can have designer resonances and prescribed permittivity and permeability, they can have engineered electromagnetic response symmetries and can localize light, creating strong concentrations of electromagnetic energy. Metamaterials can also exhibit the profound nonlocality of electromagnetic response (large metamolecules, comparable in size with the wavelength of light) and can show extreme anisotropy. Metamaterials can be tuned and switched, and can be reconfigured as arrays as well as at the individual metamolecule level.

In a historically short interval of time, from the onset of the twenty-first century, metamaterials have allowed an



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unprecedented array of fundamentally important achievements. This includes the discoveries of materials with a negative index of refraction, optical magnetism and asymmetric transmission. Metamaterials have allowed the observation of toroidal moments and optical anapoles in electrodynamics. Media with hyperbolic dispersion and an epsilon near zero were first identified in metamaterials, and they have stimulated research on topological phenomena in optics, as well as making possible the arbitrary control of light trajectories in new photonic applications such as cloaking devices. Who could have anticipated twenty years ago that the mature fields of optics and electromagnetism would be reborn? The new findings now require physics courses and textbooks on optics, electromagnetism and beyond to be re-written.

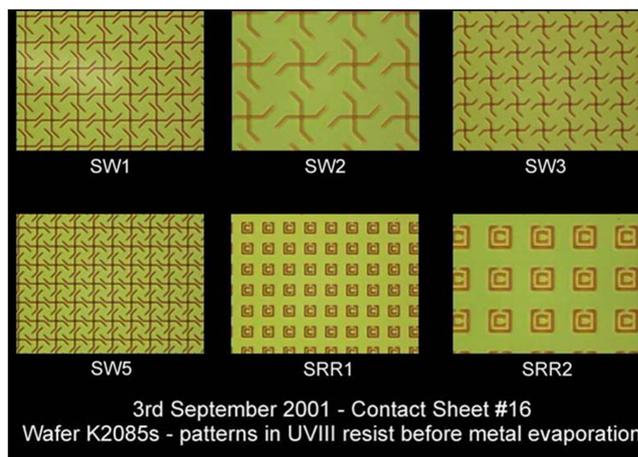
From a more practical perspective, metamaterials can control light propagation in free space and waveguides, and can be used to manipulate electromagnetic wavefronts in applications such as focusing, signal multiplexing, data processing and holography. Metamaterials are being developed that control light with external electric and magnetic signals, and they can be made extremely efficient in absorbing and harvesting it. Metamaterials can amplify and generate coherent and incoherent light and can be exploited to control light with light using record-breaking engineered nonlinearities of the novel nanostructures. They can also be used to convert and mix light frequencies and tailor the field structure at their interfaces.

In the early days of research, metamaterials were constructed by arranging sub-wavelength resonators (metamolecules) in two or three dimensional arrays. The majority of work published before 2012 explored metamaterial structures based on metals. A much wider range of material is now used to construct metamaterials. Metamaterials are now being made from metals, superconductors, dielectrics, semiconductors, graphene and carbon nanotubes, polymers and liquid crystals, conventional liquids, such as water and molten mercury, gallium, oxides and nitrides, chalcogenides, perovskites and topological insulators.

### Earlier days, from optics to microwaves and back

At the University of Southampton we came to work on what became known as the field of metamaterials at a very early stage. This activity grew from our interests in nanophotonics and chirality. In particular, our earlier studies in linear and nonlinear polarization phenomena in chiral media [4] led to a keen interest in artificial (metamaterial) chirality at the beginning stages of the discipline. Our past work on the nanophotonics of structural transformation [5] seeded our later interest in phase-change metamaterials. The highly talented Kevin MacDonald and Vassili Fedotov, who were to become key players in the future metamaterials group at the University of Southampton, joined us as my research students in 1998 and 1999 respectively, and worked on structural transformations at the nanoscale.

In November 2000, still unaware of the seminal San Diego group paper reporting negative permittivity and permeability [6], which triggered metamaterials research in many groups around the world, we submitted a paper that, in essence, described metamolecules and three-dimensional



**Figure 1.** Contact sheets of the first photonic metamaterial samples developed at the University of Southampton (N I Zheludev, D Bagnall and A Potts, 3 September 2001).

metamaterials [7]. It talked about ‘a bilayered quasiplanar metallic microstructure, comprising an ensemble of fully metallic ‘molecules’ with inductive coupling between two parts of the molecule that show strong optical rotatory power’ (figure 1). The universality and power of this idea became evident in the years that followed<sup>1</sup>.

From the very beginning of our work on structural materials, we wanted to develop functional materials for the optical part of the spectrum, and later adopted metamaterials terminology. Working on the optical part of the spectrum was a tough proposition, as it required fabrication at the sub-wavelength level, and thus nanoscale finesse was needed to develop the structures. We were very lucky to have at Southampton a fully fledged silicon foundry, and in 2001 started our journey into the exciting world of experimental research on metamaterials by fabricating two-dimensional micron-scale arrays of planar chiral ‘gammadion’ and splitting metamolecules (see figure 1). We are not aware of any other groups who were fabricating optical metamaterial nanostructures at that time. This was an important and promising step, but not yet sufficient to create truly homogeneous optical metamaterials: the available resolution was limited to about one micron. With these metamaterial structures in hand, some of which were left-right asymmetric, we decided to experimentally study diffractive polarization phenomena, a field in which no other group had previously expressed interest<sup>2</sup>. In August 2002 we submitted a paper [9] entitled ‘Optical manifestations of planar chirality’. In essence, we reported the observation of two-dimensional optical activity: that a planar chiral structure (i.e. one that cannot be

<sup>1</sup> For instance, in configurations when the coupled metallic plates forming the molecule are parallel, it shows a profound optical magnetic response, as has been exploited in a number of earlier demonstrations of optical magnetism [8].

<sup>2</sup> Such structures would eventually become known as gradient metamaterials, where metamolecules change shape or size in a periodic fashion, creating interesting diffraction effects. In particular, in 2011 the Capasso group provided a systematic study of diffraction on arrays of polarization-sensitive metamolecules, which used the same V-shaped constitutive blocks as our gammadion arrays and became known as photonic metasurfaces [10].

completely superimposed over its mirror image) can affect the polarization state of diffracted light. This observation showed that polarization rotation changed its sign for two enantiomeric forms of the media. The fundamental role that planar chirality (not to be confused with the three-dimensional chirality that underpins the conventional optical activity effect) plays in polarization phenomena was identified in this paper. This work was sponsored by what was arguably the first metamaterial research grant in Europe, awarded to me by the Science and Engineering Research Council (UK). Beginning on 1 October 2002, the grant was titled ‘Optical manifestations of planar chirality,’ GR/S00958/01. Around that time we also met and started a fruitful collaboration with a theorist from Kharkov, Ukraine, Professor Sergei Prosvirnin, who came to visit our group a few times in the years to come.

On the same planar arrays of chiral gammadions we saw that in a diffraction experiment the polarization eigenstates changed if the direction of light propagation through the structure was reversed [9]. This result initiated our studies of propagation asymmetries with planar chiral metamaterials. We soon reported the ‘asymmetric transmission effect’, where the overall intensity measured upon the transmission of a circularly polarized wave through a planar chiral metamaterial depended on the direction of transmission [11]. This was a shocking discovery for many, as normally one would expect the optical properties of a medium in the reverse direction to be exactly the same in the absence of a magnetic field, according to the Lorentz reciprocity consideration. In fact, the observed phenomenon is underpinned by a polarization-conversion process, which is allowed by reciprocity. The observed effect is fundamental, as it adds to the pantheon of basic polarization effects. It is distinct from conventional anisotropy with orthogonal linear eigenstates, and from the conventional gyrotropy of bulk chiral media and the Faraday effect, where the eigenstates are a pair of counter-rotating elliptical states. Indeed, the eigenstates of a lossy anisotropic planar chiral structure are two co-rotating elliptical polarizations. In the following years, we also demonstrated this effect in plasmonic nanostructures in the optical [12, 13] and THz parts of the spectrum [14].

On 30 October 2005 disaster struck: a fire destroyed our fabrication platform, the Southampton Silicon Foundry, and many photonic labs. Our ambition to push for photonic metamaterials became considerably more challenging without direct access to the sophisticated fabrication facility. However, we were able to keep working on metamaterials, as in 2003 we secured funding to set up a microwave characterization facility located in a different building. Microwave capability allows relatively simple measurements and cheap samples (in most cases fabricated by printed circuit board technology). Diverting to microwave research turned out to be a wise step at a troubling time for our nanofabrication capability: the fire slowed our photonics work, but allowed us to start a number of new research projects.

During this period of time in 2005, we demonstrated a metamaterial mirror that resonantly ‘amplifies’ losses in the dielectric layer sandwiched between the metamaterial and

metal plate [15]. Later this became known as a ‘perfect absorber’. This device showed nearly 40 dB resonant suppression of reflectivity, attributed to the high concentration of fields near the metamaterial, and remaining the best performance for many years. We identified applications of high-absorbing metamaterials in the spectroscopy of weak lines, and improvements to the sensitivity of photodetectors. We also demonstrated that microwave structures can be used as ‘magnetic walls’ that repel magnetic fields, in a similar way in which a static magnetic field is repelled from superconductors by the Meissner effect. Such a mirror reflects like a conventional mirror without diffraction, but shows no phase change with respect to the incident wave [16]. In a few years, we provided a demonstration of the magnetic wall in the optical part of the spectrum [17]. In 2005 we also found a way of making metamaterials invisible, and experimentally demonstrated a microwave metamaterial that at some frequencies contributes no transmission losses or phase delay [15]. We called this an ‘invisible metallic structure,’ or ‘invisible metal.’<sup>3</sup>

Deprived by the fire of our own nanofabrication resource, we started a very important collaboration with Dr Yifang Chen from the Rutherford Appleton Laboratory in Didcot (now at Fudan University, China). Due to the Didcot fabrication facility limitations, we were not able to work with gold and silver—good plasmonic materials used by other groups—but instead embarked on the very early use of aluminium as a metal component in plasmonic metamaterial nanostructures. At this facility, since 2005 we have also been experimenting with soft lithography and nanoimprinting to develop high-throughput metamaterial fabrication techniques [19].

The experimental realization of chiral microwave metamaterials with electromagnetic coupling predicted in our earlier work [9] was reported by our group in 2006 [20]. In these structures, we found extremely strong polarization rotatory power; in terms of rotatory power per sample thickness of one wavelength, the bilayered structure rotated five orders of magnitude more strongly than a gyrotropic crystal of quartz in the visible spectrum. Moreover, we saw clear evidence of negative refraction in the metamaterials by identifying a range of frequencies where the phase velocity and group velocities of light had opposite signs for one of the circular polarizations. A detailed analysis of the observed effect was developed a few years later [21]. A year after this we demonstrated a three-dimensional chiral photonic metamaterial by fabricating a multi-layer nanostructure [22]. Multi-layered structures would later be referred to as ‘stereo metamaterials’. We were able to achieve nanoscale mutual alignment of individual layers and create large arrays of metamolecules only 700 nm × 700 nm in size. In the visible and near-IR spectral ranges, the metamaterial exhibited polarization rotatory power of up to 2500 degrees mm<sup>-1</sup>, showed relatively low losses and negligible circular dichroism.

<sup>3</sup> Concurrently, a similar scattering cancellation idea was being theoretically discussed by the Engheta group [18].

Nikitas Papisimakis and then Eric Plum joined the metamaterials group at Southampton as my research students in 2005 and 2006, respectively, and started intense work in exciting new directions of metamaterials research: trapped-modes, slow light, collective resonances and artificial chirality. In the years to come, they will become key collaborators and players in our metamaterials research programmes.

Metamaterials have also helped us to understand that the classical phenomenon of optical activity can be observed in artificial planar media, which exhibit neither 3D nor 2D chirality [23]. We experimentally observed what we called ‘extrinsic optical activity’ in the microwave and optical parts of the spectrum by passing light at oblique incidence through regular arrays of nonchiral subwavelength metamolecules. We saw strong circular dichroism and birefringence that was indistinguishable from that of chiral three-dimensional media, but which arose from the mutual orientation of the metamaterial and the incident beam. As an extension of this work, we recently looked at the reflective effects of chiral metasurfaces [24]. We demonstrated a type of mirror reflecting one circular polarization without changing its handedness, while absorbing the other [25]. The polarization-preserving mirror consists of a planar metasurface and a conventional mirror spaced by a fraction of the wavelength from the metasurface. Such mirrors should enable circularly polarized lasers and Fabry–Perot cavities with enhanced tunability, gyroscopic applications and polarization-sensitive detectors of electromagnetic waves, and can be used to enhance the spectroscopies of chiral media. Furthermore, we also discovered that the directionally asymmetric transmission of circularly polarized waves occurs at oblique incidence on structures that are not planar chiral [26, 27].

As a continuation of earlier works of the 1980s on microwave frequency selective surfaces, a practically important direction emerged in metamaterials research around 2006, as scientists looked to realize narrow, low-loss resonances and strongly dispersive behaviour in planar, two-dimensional structures of sub-wavelength thickness by using interference effects that suppress radiation leakage. These advances would greatly expand the metamaterial playground to encompass sensors, compact delay lines and coherent light-emitting devices. In 2007, we observed that an array of asymmetrically split ring metamolecules exhibits a classical analogue of the narrow resonances observed in electromagnetically induced transparency and Fano-type resonances [28, 29]. In the years that followed, the asymmetric split ring design would become one of the most popular metamaterial patterns, due to the manifestation of high-quality resonance modes with easy-to-control quality factors. We then studied classical analogues of the electromagnetically induced transparency (EIT) effect in detail and showed that pulses propagating through such metamaterials experience considerable delay. The thickness of the structure along the direction of wave propagation is much smaller than the wavelength, which allows the successive stacking of multiple metamaterial slabs leading to high transmission and broadband slow light [30, 31].

At this point we realized that metamaterials research was a much broader paradigm than just controlling permeability, permittivity and nonlocality, and we began thinking about metamaterials in a much wider context [2], stating: ‘The next stage of this technological revolution will be the development of active, controllable, and nonlinear metamaterials surpassing natural media as platforms for optical data processing and quantum information applications. Metamaterials are expected to have an impact across the entire range of technologies where electromagnetic radiation is used, and will provide a flexible platform for modelling and mimicking fundamental physical effects as diverse as superconductivity and cosmology, and for templating electromagnetic landscapes to facilitate observations of phenomena that would otherwise be difficult to detect.’ In the years that followed, we witnessed the realization of these predictions.

In 2008 we introduced a lasing spaser, which was a further development of the spaser concept. We showed that by combining metamaterial and spaser ideas one can create a narrow diversion, coherent source of electromagnetic radiation that is fuelled by plasmonic oscillations. We argued that a two-dimensional array of plasmonic resonators, supporting high-quality factors and coherent current excitations across the array, can act as a planar source of spatially and temporally coherent radiation (see below for more details on ‘coherent’ metamaterials). In the years that followed, this idea would be realized by a number of groups [32]. Recently, I discovered that Richard Feynman, in the days before metamaterials, had essentially proposed this for an array of nanoscale coherent emitters: ‘Consider ... a piece of material in which we make little coils and condensers 1000 or 10 000 angstroms in a circuit ... over a large area, with little antennas sticking out at the other end ... to emit light from a whole set of antennas ... ?’[1].

Our work in the field of light-emitting metamaterials included the observation that hybridizing semiconductor quantum dots with a plasmonic metamaterial leads to a multifold intensity increase and the narrowing of the photoluminescence spectrum. This manifestation of the cavity Purcell effect can also be controlled by the design of the nanostructure [33]. This observation was a step towards understanding loss compensation in plasmonic metamaterials, and developing metamaterial enhancement for gain media.

From 2004 until the end of 2009, our metamaterial and nanophotonic activities at the University of Southampton were sponsored predominantly by my EPSRC Nanophotonics Portfolio grant EP/C511786/1. Recognition of metamaterial research as a hugely important direction has grown in the UK physics community, and in 2010 the Engineering and Physical Sciences Research Council (UK) awarded my group with a large programme grant titled ‘Nanostructured photonic metamaterials,’ EP/G060363/1.

At the end of 2006, I moved my research labs and entire research group from the School of Physics at the University of Southampton to the world-famous Southampton Optoelectronics Research Centre (ORC), where I was appointed Deputy Director (physics). From that time, I had the privilege of working with the ORC Director, Professor Sir David Payne,



**Figure 2.** University of Southampton Mountbatten Building, which hosts the Optoelectronics Research Centre and the Centre for Photonic Metamaterials, and stands on the site of the Southampton Silicon Foundry; it is arguably the only building in the world covered with a metamaterial design. The inset (top left) shows a fragment of the planar-chiral fractal metamaterial pattern developed within the first metamaterial research grant ‘Optical manifestations of planar chirality’ awarded to NIZ in 2002. The pattern was developed by A Potts, D Bagnall and N I Zheludev, and was fabricated on a silicon wafer at the Southampton Silicon Foundry shortly before fire destroyed it. Its design was the inspiration for the pattern on the building.

FRS, and the ORC Deputy Director (technology), Professor David Richardson. In my new post, I succeeded Professor David Hanna, FRS, a famous British laser scientist who, in 1991, had played a crucial role in my initial appointment at Southampton, and had recently retired. The diverse environment of the ORC had a very positive and stimulating impact on our work, and in particular helped us to align it with the needs of laser and optical telecommunication technology. Several people at the ORC became active contributors to our programme, most notably Professor Robert Eason and Professor Dan Hewak, experts in nonlinear optics and photonic materials. This move to the ORC has profoundly and positively affected the destiny of metamaterials research at Southampton, and is now a primary research direction of the ORC and the University of Southampton.

Through the late 2000s, the international metamaterial community grew rapidly, attracting bright people from various disciplines such as microwave engineering, solid state physics, photonics, materials science, nanotechnology and computational science to this new synthetic field of research. A number of new conferences emerged. The Optical Society of America launched a series of conferences on ‘Photonic Metamaterials and Plasmonics’ located in The Bahamas (2006), Jackson Hole, WY (2007), Rochester, NY (2008), and Tucson, AZ (2010). I worked on the Jackson Hole conference with Professor Vlad Shalaev from Purdue University and was also engaged in conferences with learning societies in Europe. On 18 December 2002, Professor Sergei Tretyakov (Aalto University, Finland)

initiated a meeting in Brussels to lobby for a European network of excellence sponsored by the European Union. It was attended by a few enthusiasts of the discipline, including Professor Didier Lippens (France), Professor Ferran Martín (Spain), Dr Nigel Johnson (UK), Professor Andre de Lustrac (France), Professor Tomasz Szoplik (Poland), Professor Nikolay Zheludev (UK), Dr Alex Schuchinsky (UK) and Dr F Javier García de Abajo (Spain). The meeting was also attended by Dr Gustav Kalbe, a representative of the EU funding body. Eventually, this led to the formation of the ‘Metamorphose Network of Excellence’ in 2003. In 2008, this became ‘The Virtual Institute for Artificial Electromagnetic Materials and Metamaterials’ (‘Metamorphose VI’). The most important legacy of this development was the creation of the ‘Metamaterials Congress,’ a regular conference event on our calendar, recurring yearly from 2007 and held consecutively in Rome, Pamplona, London, Karlsruhe, Barcelona, St Petersburg, Bordeaux, Copenhagen, Oxford, Crete and Marseille. I chaired the programme committee of the first congress in Rome. In 2008 Professor Said Zouhdi (France) single-handedly started a new nanophotonics and metamaterials conference called META that was held consecutively in Marrakesh, Cairo, Paris, Dubai, Singapore, New York, Malaga and Seoul, and for which I co-chaired the Singaporean conference with Professor Zouhdi. SPIE also began a series of annual conferences on metamaterials, one in the USA and another in Europe, now co-chaired by Kevin MacDonald. Jointly with Professor Mikhail Noginov and Nader Engheta I have, since 2010, also co-chaired the USA branch of this conference. Currently titled ‘Metamaterials, metadevices, and metasystems,’ the conference is traditionally held in San Diego, CA. For our Southampton group, the most significant conference development was the creation of ‘NANOMETA’ in 2007. It is now a flagship European Physical Society international meeting at the crossroads of nanophotonics and metamaterials. NANOMETA is run biennially in Seefeld, Austria (2007, 2009, 2011, 2013, 2015, 2017, see: [www.nanometa.org](http://www.nanometa.org)). I proposed the first meeting and organized it jointly with Professor Ekmel Ozbay (Turkey), and since 2013 we have co-chaired the meeting with Professor Harald Giessen (Germany) [34].

### Developing metamaterials technology: switching and tunability

With the award of the dedicated photonic metamaterials programme grant, the Centre for Photonic Metamaterials at the University of Southampton ([www.nanophotonics.org.uk](http://www.nanophotonics.org.uk)) was formed with me as director (see figure 3). Kevin MacDonald was appointed research manager of the programme and the new centre. Professors Robert Eason (ORC), Peter DeGroot (School of Physics and Astronomy), Janne Ruostekoski (School of Mathematics) and Dan Hewak (ORC) joined the programme as co-investigators. Dr Vassili Fedotov contributed to the programme in the capacity of EPSRC Career Acceleration Fellow, and subsequently joined the faculty at Southampton. Simultaneously, Eric Plum and Nikitas Papanikolaou received advanced fellowships and shortly afterwards

became assistant professors at Southampton. This was made possible in 2010 with help from The Leverhulme Trust programme on 'Embedding the new discipline of metamaterials into UK universities' (jointly held with the programme PI, Prof. Sir John Pendry at Imperial College). A substantial, stable critical mass of research in metamaterials at the University of Southampton was formed with many academics, researchers and students involved. A prominent international theorist, Professor F Javier García de Abajo, spent nearly two sabbatical years with us. We had a constant flow of long-term international visiting researchers to the centre, most notably from Japan. A strong collaborative link with Professor Din Ping Tsai from the National Taiwan University had become a constant feature of our work, and the source of many results and much inspiration. The programme grant was a strong catalyser for our research, and during its five-year period, we were able to attract more than twenty additional grants and contracts related to metamaterials technology.

The Southampton silicon foundry, which had been destroyed by fire in 2005, was resurrected at the university in 2010 as the Mountbatten Complex—a dedicated new building with 1500 square metres of high-class cleanrooms. We were honoured to see that the award-winning building featured a full-façade glass surface emblazoned with our metamaterial design (see figure 2). It would soon also host the Southampton Nanofabrication Centre, a state-of-the-art facility for microfabrication and high-spec nanofabrication, and the Southampton Photonics Foundry, a hub for innovation in photonic science and technology. This fabrication facility would provide unique processes for manipulating a broad spectrum of photonic materials and devices such as novel glass, integrated photonics, and advanced optical fibres, for use in applications such as biophotonics, nanophotonics, and metamaterials. However, more time was needed to make it fully operational. At that stage a new collaboration emerged that helped us during the difficult years of deprivation from an in-house fabrication capability. The group of Professor Di Fabrizio, which is a world leader in innovative nanofabrication from the Italian Institute of Technology, worked with us on complex nanostructures for three years between 2010 and 2012.

While the majority of the research community was still exploring the intriguing opportunities that negative index materials could provide and transformation optics could deliver, our vision for the programme granted in 2010 was to develop a new generation of switchable and active nanostructured photonic media for telecoms, energy, light generation, imaging, lithography, data storage, sensing, security and defense applications. The main methodological paradigm for the programme was to achieve new functionalities by developing hybrid photonic metamaterials.

Hybridization is a powerful approach, in particular for developing highly nonlinear and switchable metamaterials by harnessing resonant local field enhancement and plasmonic resonances. For instance, the hybridization of a nanoscale 'feutre' of single-walled carbon nanotubes with plasmonic metamaterials creates a photonic medium with exceptionally strong ultrafast nonlinearity [35]. This behaviour is

underpinned by coupling the nanotube excitonic response to weakly radiating Fano-type resonant plasmonic modes that can be tailored by metamaterial design. The strongest response with a picosecond relaxation time was achieved when the carbon nanotube resonance frequency matched that of the plasmonic mode. In the past, we used powerful and complex nanosecond, picosecond and femtosecond lasers for measuring the nonlinear properties of materials. However, when studying carbon nanotube metamaterial hybrids, it was sufficient to use only a few milliwatts of power from a spectrally filtered, broadband quasi-continuous wave (CW) supercontinuum source<sup>4</sup>, which is an indication of the strength of the nonlinear response. With the rise of graphene, we rapidly recognized that photonic metamaterials could exhibit extraordinary sensitivity to the presence of a single atomic layer of graphene on its surface [36]. The highly broadband, but relatively weak nonlinearity of graphene can be enhanced about twenty times by placing it on a resonant plasmonic metamaterial, which is another demonstration of the hybridization concept [37].

I had studied the ultrafast optical nonlinearities of metals [38–40] for a few years before we originally embarked on research into metamaterials. It was clear to me that the metallic framework of the metamaterial itself should have a substantial nonlinear response, even surpassing that of unstructured metallic films. Indeed, we have found that the third-order, two-photon-absorption optical nonlinearity of gold films can be greatly enhanced, and that even its sign can be controlled by metamaterial nanostructuring. This discovery offers applications in ultrafast optical limiters, saturable absorbers and terahertz-bandwidth all-optical gates [41].

This brings me to the story that, in my view, epitomizes the power of the metamaterial paradigm. In 1979, as a student, I was charged with observing the effect of nonlinear optical activity theoretically described by my mentor, Prof. Sergei Akhmanov in 1967. I spent a lot of effort and displayed inventiveness in detecting the minute dependence of the polarization rotatory power on the light intensity in a crystal of lithium iodate. This was a tough job. Thirty-three years later, thanks to our development of metamaterials, which exhibit strong optical activity and nonlinearity in the constituting metal, the observation of a nonlinear optical activity has become trivial: we reported that a plasmonic metamaterial exhibits nonlinear optical activity that is 30 million times stronger than lithium iodate crystals, thus transforming this fundamental phenomenon of polarization in nonlinear optics—observed back in 1979—from an esoteric phenomenon into a major effect encountered in nonlinear plasmonics, with potential for practical applications [42].

Using superconductors was one of the most intriguing opportunities for making low-loss tunable metamaterials. Indeed, superconductivity is very sensitive to heat, magnetic

<sup>4</sup> Supercontinuum lasers are now widely used in nanophotonics and metamaterials research. Our research used one of the first commercial models, developed by another close colleague Dr Anatoly Grudinin, who is a former ORC researcher, founder and CEO of Fianium Ltd, which is now a multi-million-pound laser business in Southampton that recently became a part of NKT Photonics.

field and current stress, which gives researchers a number of new degrees of freedom. In 2009 we decided to set up a characterization rig for superconducting metamaterials, and elected to work at the spectral band around 100 GHz in the millimetre-wave part of the spectrum, using a recently developed new generation of high-frequency vector network analysers (HP/Agilent). At these frequencies, we were able to work with both high-temperature, cuprate superconductors, and with conventional superconducting niobium. Moreover, quasi-optical techniques, optical cryostats, as well as other components to which we were well accustomed, were suitable for these frequencies. What excited us was that a negative dielectric constant and dominant kinetic resistance made superconductors a natural, broadband, plasmonic media [43, 44]. In our earlier experiments, we targeted losses as the main limiting factor in the use of metamaterials in practical applications. While radiation losses may be controlled by design, Joule losses are inherent to the metamaterial structures. Superconducting metamaterials are an exception to this, since Joule losses can be uniquely controlled with temperature across a very wide range. We put this concept to use by demonstrating temperature-dependent transmission of the hole array in a high-temperature (high-Tc) superconducting cuprate film, measured in the millimetre-wave part of the spectrum [43] and metamaterials, supporting sub-radiant Fano-type resonances [45]. At that point, only a handful of groups were working on superconducting metamaterials and we benefited from discussion and collaborations with Professor Steven Anlage at the University of Maryland, who came to visit us on a few occasions. Our further work included the demonstration of a frequency-selective, energy-harvesting, sub-THz superconducting bolometer using ‘coherent metamaterials’ [46], and a sub-THz electro-optical modulator based on a niobium superconducting metamaterial, where current passing through a network of superconducting metamolecules controls the state of superconductivity [47]. We also explored the idea of flux-exclusion quantum metamaterials [48]. However, perhaps the most relevant illustration of what it is possible to achieve with a highly sensitive state of superconductivity is our superconducting, sub-THz nonlinear metamaterial that operates in the CW regime. This device exhibited a record-breaking resonant, third-order nonlinearity with an effective  $n^2 = 10 \text{ cm}^2 \text{ W}^{-1}$  [49]. We designed this metamaterial in such a way that the heat of absorbed electromagnetic radiation was released in nanoscale constrictions of the superconducting metamolecules, suppressing superconductivity and higher powers. Using this mechanism of nonlinearity it was possible to achieve a profound nonlinear response in the structure at a power level of only  $500 \mu\text{W cm}^{-2}$ .

The cubic optical nonlinearities that are responsible for the light-induced change of refraction and absorption allow the control of light with light in a volatile fashion: a switched medium relaxes to its initial state after excitation is withdrawn. Nonvolatile switching, which remains in the absence of the excitation source, requires a material with ‘memory.’ From 1998 onward, we worked on nanoscale structural transformations as a source of optical nonlinearity and

nonvolatile switching [5], with keen interest in using them for photonic technologies. We realized that phase-change functionality could provide a way of shrinking optical switching devices to the nanoscale [50], and later that it could be used to control the individual molecules of metamaterials [51]. Indeed, the modulation of a signal requires that its amplitude or phase be changed substantially by a control signal, through a change in either propagation losses or propagation delays. The only way to overcome the lack of propagation distance in nanoscale elements is to use nanometre-scale materials that can undergo a very strong change in absorption or refraction in response to external control. To accomplish this via a material’s refractive index, the real and imaginary parts would need to change on the order of unity. Large changes in the optical properties of a material can be achieved by providing the energy needed to change it from one structural phase to another. This energy ‘price’ is typically of the order of the characteristic phonon energy per atom. The energy of only a handful of photons could be enough to convert an entire nanoparticle or metamolecule made of phase-change material from one phase to another.

There are two main types of structural phase-change transformation that are particularly useful in photonic metamaterials: ‘ovonic’ phase-change and ‘phase-change coexistence.’ In the latter, a nanoparticle or the film of a material may have two different structural phases of matter that coexist simultaneously, with the balance between them controlled by temperature or an external supply of energy. An early example from our work is the nanoscale film of liquid gallium existing at the interface between solid gallium and silica glass [52]. Similar coexistence arrangements may be found in gallium nanoparticles where, depending on the temperature, solid-liquid or solid-solid core-shell particles made of different structural phases are formed [53]. During the pre-metamaterial era we extensively studied such gallium films and nanoparticles as platforms for all-optical and free-electron-driven switching and memory elements [5, 54–56]. Today, the metamaterial terminology is used widely, and structural materials consisting of arrays of nanoparticles are commonly referred to as metamaterials. In 2002, we demonstrated all-optical switching in gallium nanoparticles [57], and in 2003 we demonstrated the memory effect in this metamaterial [58]. In 2006 we demonstrated nanocomposite metamaterials created by the grain boundary penetration of gallium into an aluminium film. These composite films form mirror-like interfaces with silica and show an exceptionally broadband phase-transition-based nonlinear response to optical excitation [59]. Recently, we demonstrated an optically switchable, hybrid, plasmonic, gallium-based metasurface, in which a reversible light-induced transition between solid and liquid phases, occurring in a confined nanoscale surface layer of the metal, drives significant changes in reflectivity and absorption [60]. The metasurface architecture resonantly enhances the metal’s ‘active plasmonic’ phase-change nonlinearity by an order of magnitude, offering high-contrast all-optical switching in the near-infrared range at low  $\mu\text{W } \mu\text{m}^{-2}$  excitation intensities.

The ‘ovonic’ phase change transformation type is most often seen in chalcogenide semiconductor materials, and happens as a gradual destruction of the crystalline lattice, converting the material into the amorphous phase. By contrast, the transition from the amorphous state to the crystalline one is usually abrupt. Phase-change in chalcogenide materials, such as germanium antimony telluride (GST) and gallium lanthanum sulfide (GLS), is a very robust process that is used in re-writable optical and electronic data storage. In 2010 we demonstrated hybrid-plasmonic electro-optic switching in chalcogenides. To accomplish this, we exploited the frequency shift of a narrow-band Fano resonance mode in a plasmonic planar metamaterial, induced by a change in the dielectric properties of an adjacent chalcogenide glass layer [61]. The all-optical switch combines the chalcogenide glass phase-change medium with nanostructured, plasmonic metamaterials to yield a high-contrast, large-area, reversible switching solution [62].

Importantly, phase-change is a technology that can work for metamolecule-level switching, providing a new platform for creating optical components that are written, erased and rewritten as two-dimensional binary or greyscale patterns into a film of chalcogenide glass using tailored trains of femtosecond pulses. By combining germanium-antimony-tellurium-based films with a diffraction-limited-resolution optical writing process, we demonstrated that reconfigurable bichromatic and multifocus Fresnel zone plates, superoscillatory lenses with subwavelength focus, grayscale holograms, reconfigurable photo-masks and dielectric metamaterials with on-demand resonances can be created with this technology [63, 64]. Today, phase-change is widely seen as a key technology for nanophotonic and metamaterial switching and memory, and many leading nanophotonic research groups run projects developing phase-change technology.

More and more, we focused our work not only on developing metamaterials, in which the change of optical response could be achieved across the entire volume of the metamaterial, but also on those in which the ‘on-demand’ control—change at any given point in space and time—of individual metamolecules would be possible [3]. Analogous with electronic random access memory, we called them ‘randomly accessible metamaterials.’ These metamaterials would not only allow for the modulation of light’s intensity or phase, but would offer full, active control of the wavefront of electromagnetic radiation, tailoring of the near field, and, ultimately, multichannel data processing. Developing randomly accessible photonic metamaterials is a challenge: the metamolecules must be subwavelength optical switches with a physical volume of about  $10^{-19} \text{ m}^3$ . To have an impact on telecommunications technologies, such switches must also be fast and energy-efficient. At Southampton we were looking for such technologies that could deliver not only metamolecule-level switching controlled by electric or magnetic signals, but also switching with light [3]. One such light-sensitive process is the phase-change technology described above.

Some exceptional opportunities have also been provided by nano-optomechanics, which take advantage of the

changing balance of forces at the nanoscale. At the sub-micrometre dimensions of metamolecules, Coulomb, Ampère and Lorentz forces, compete with elastic forces and can thus be used to reconfigure the shape of individual metamolecules, or their mutual arrangement. We have identified that nano-membranes are the ideal platform for such nano-optomechanical reconfigurable metamaterials [65]. Metamaterials on structured membranes can be reconfigured thermoelastically [66], mechanically [67], electrically [68] and magnetically [69]. They can also be reconfigured by the light-induced forces that occur between elements of illuminated metamolecules. We have shown this in both plasmonic [70] and dielectric metamaterial nanostructures [71]. It is now clear that the nonlinear, switching, electro-optical and magneto-optical characteristics of nano-optomechanical metamaterials can surpass those of natural media by orders of magnitude. For instance, the electro-optic coefficient of a reconfigurable metamaterial exceeds that of a reference lithium niobate crystal by five orders of magnitude [68]. Nano-optomechanical metamaterials also show exceptionally strong optical nonlinearities. Dielectric metamaterial nanostructures can modulate light by light at 150 MHz with a nonlinear coefficient of  $\beta \approx 7 \cdot 10^{-5} \text{ m W}^{-1}$  [71], while plasmonic nano-optomechanical metamaterials modulating at a frequency of 1 MHz show  $\beta \approx 5 \cdot 10^{-3} \text{ m W}^{-1}$  [70]. Such a high level of nonlinearity allows for cross-wavelength, all-optical signal modulation with conventional telecom diode lasers of only a few milliwatts. Importantly, nanoscale metamaterial building blocks can be moved very fast, potentially offering gigahertz bandwidth switching, with modulation at a hundred MHz already having been achieved [71]. The first generation of randomly accessible, reconfigurable photonic metasurfaces on nano-membranes, providing control in a single spatial dimension, has also been realized [65]. Such metasurfaces can function as re-focusable lenses or dynamic diffraction gratings [72–74]. I believe that soon we will be able to see the practical demonstration of an intrinsically bistable metamaterial, driven by resonant optomechanical forces. Being inherently free of Joule losses, we expect this to exhibit optical bistability at intensity levels of less than  $\text{mW } \mu\text{m}^{-2}$  [75].

Another emerging technology for controlling and switching optical properties in metamaterials is ‘coherent control’ [76]. In 2012 we realized that a highly absorbing plasmonic metamaterial film of subwavelength thickness that is placed at the node of a standing wave, and formed by counter-propagating control and signal waves, will see zero electric field and thus will not absorb light. Any change in the phase or intensity of the control wave will distort the standing wave pattern and destroy the regime of zero absorption. This method can be used to control various optical effects, such as diffraction [77], optical activity and birefringence [78], as well as nonlinear interactions [79]. This phenomenon underpins various forms of optical switching [80] with only a few photons [81], and with a modulation bandwidth up to 100 THz using plasmonic and dielectric metamaterial absorbers [82–84]. This presents powerful opportunities for laser spectroscopy [85–87] and data handling in locally coherent networks, which are increasingly becoming a part of

mainstream telecommunications development. The ability to control the wavefront of light is fundamental to the focusing and redistribution of light, enabling many applications from imaging to spectroscopy. Coherent interactions can also be used for the two-dimensional control of light with light on highly absorbing plasmonic metasurfaces. In 2015 we demonstrated this by performing 2D all-optical logical operations (AND, XOR and OR) and image recognition and processing [88, 89], offering potential solutions for all-optical data processing in multi-core fibre networks [90]. Our approach offers diffraction-limited image processing, potentially at arbitrarily low intensity levels, and with 100 THz bandwidth, thus promising new applications in space-division multiplexing, adaptive optics, image correction, processing and recognition, 2D binary optical data processing and reconfigurable optical devices.

The interaction of free-electron beams with nanostructures has fascinated me for many years. We studied phase coexistence in gallium nanoparticles controlled by electron excitation [91]. Concurrently with Albert Polman's group in AMOLF, but a few days earlier, we published on the generation of travelling surface plasmon waves by free-electron impact [92]. We also demonstrated amplification of the evanescent field of free electrons [93]. We developed a nanoscale analogue of a free electron light source by demonstrating that the passage of a free-electron beam through a nanohole in a periodically layered metal-dielectric structure (nano undulator) creates a new type of tunable, nanoscale radiation source that we termed a 'light well' [94]. The study of light generation in metamaterials has become a logical extension of this work, and in 2012 we showed experimentally that the energy from a highly localized free-electron-beam excitation can be converted via a planar plasmonic metamaterial to a low-divergence free-space light beam [95]. This emission, which emanates from a collectively oscillating coupled metamolecule nanoantenna ensemble much larger in size than the initial excitation, is distinctly different from cathodoluminescence and bears some similarity with laser light. This offered a novel, flexible paradigm for the development of scalable, threshold-free light sources. Recently, we developed a direction and mode-tunable holographic free-electron light source based on the injection of free electrons into gradient metasurfaces [96].

Our 2011 paper on changing the perceived colour of gold through metamaterial patterning, without violation or employing any form of chemical modification, attracted huge interest from the general public, watch manufacturers, jewellery and luxury goods industries. We showed that on a continuous gold film the colouring effect may be underpinned by plasmonic Joule losses in the constituent elements of the patterns (dubbed 'intaglio' and 'bas relief' metamaterials to distinguish indented and raised structures respectively) [97, 98]. This colouring method has the advantage of maintaining the integrity of metal surfaces and is well suited to high-throughput fabrication via techniques such as nano-imprinting.

It took some time for the metamaterials community to realize that the many exotic and useful functionalities of metamaterials could be realized without using metals as

building materials for metamolecules, and that results similar to those of plasmonic devices could be obtained with all-dielectric structures based on high-index dielectrics. We had interest for this topic while still in its earlier stages, and in 2003 developed an effective medium theory of a metasurface, a closely packed nanoparticle film of spheroids and nanoshells with arbitrary dielectric properties [99]. In 2005 we developed techniques for fabricating dielectric metamaterials for the optical part of the spectrum and studied their polarization properties [19]. This included the fabrication of various chiral shapes in silicon with a feature of about 50 nm. We also fabricated planar chiral metamaterials in a dielectric layer of hydrogen silsesquioxane resist and used room-temperature nanoimprint lithography to replicate metallic planar chiral structures by metallization and lift-off processes.

To continue our work on dielectric metamaterials, in 2013 we experimentally demonstrated a fully dielectric (silicon) metasurface that exhibits a magnetic resonance at optical frequencies, founded upon excitation by the incident light of anti-parallel displacement currents, in metamolecules comprising pairs of dissimilar dielectric nanobars, showing that in principle, strong, lossless, resonant responses are possible anywhere in the optical spectral range [100, 101]. Moreover, we saw the suppression of reflectivity caused by the interference of magnetic and electric dipole responses, which was referred to as the Huygens surface behaviour. The same year, we introduced a dielectric photonic nonlinear metamaterial that is inherently free of Joule losses [75]. We predicted that it would exhibit a giant nonlinear optical response and nonlinear asymmetric transmission with a forward/backward optical extinction driven by resonant optomechanical forces in a structure comprised of dissimilar dielectric nanobars on a flexible membrane. As we have already noted above, we experimentally demonstrated highly nonlinear, dielectric, optomechanical metamaterials two years later [71], thereby offering a path to fast, compact and energy-efficient all-optical metadevices. In 2016 we demonstrated an all-dielectric phase-change reconfigurable metasurface [102]. We harnessed nonvolatile, amorphous-crystalline transitions in the chalcogenide phase-change medium germanium antimony telluride to realize optically switchable, all-dielectric metamaterials that provided reflectivity and transmission switching contrast ratios of up to 5:1 at visible/near-infrared wavelengths selected by design. We also demonstrated that dielectric metamaterials can be written, erased and rewritten as two-dimensional binary or greyscale patterns into a nanoscale film of phase-change material by inducing a refractive-index-changing phase transition with tailored trains of femtosecond pulses [51]. In collaboration with Professor Brian Hayden from Southampton's Department of Chemistry—a pioneer and global leader in the field of 'high-throughput materials discovery'—and colleagues in Singapore, we continue to explore new plasmonic and dielectric materials including chalcogenides, superconductors, topological insulators [71] and perovskites [103].

The school of mathematics theory group of Professor Janne Ruostekoski and Dr Stewart Jenkins (another Leverhulme fellow on my grant) provided considerable input in

several aspects of our programme by developing microscopic and quantum mechanical descriptions of the collective response of metamaterials [95, 104–108].

Metamaterials are also extremely powerful platforms for fostering the observation of new physical phenomena. The asymmetric transmission effect discussed above was one of the examples. Another unusual phenomenon, which has never been reported in natural solid or liquid media in the entire history of optics, and which was possible to ‘engineer’ in metamaterials, was the dependence of the absorption line width on sample size<sup>5</sup>. In 2010 we reported on a metamaterial in which the spectral line collapses with an increasing number of metamolecules: bigger samples had narrower lines [109]. We observed this effect at microwave, terahertz and optical frequencies, and linked it to the suppression of radiation losses in periodic arrays of strongly interacting metamolecules (collective subradiance). We later referred to this as an optical analogue of the famous Mössbauer effect, where a narrow resonance for nuclear absorption results from the recoil being delivered to the entire crystal lattice rather than to the nucleus alone.

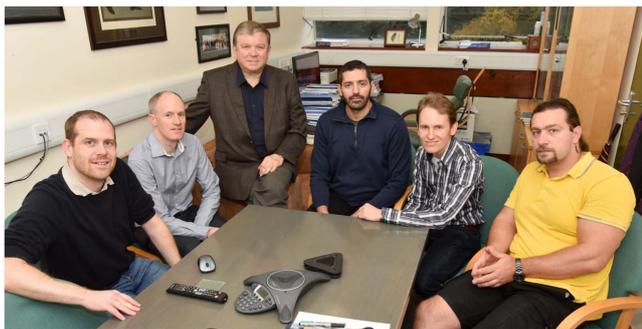
Spectral collapse is relevant to the concept of ‘coherent’ and ‘incoherent’ metamaterials, which we introduced in 2009 by experimentally demonstrating two distinctly different types of resonant metamaterial responses upon disordering the metamaterial lattice [110]. In the case of a coherent metamaterial, a regular ensemble of metamolecules exhibits a collective narrow-band response that becomes broader and eventually disappears with increasing disorder. In an incoherent metamaterial, the disorder has little effect on the structure’s resonant properties. Furthermore, we observed that in metamaterials with strongly interacting metamolecules, positional disorder can lead to light localization [111].

We also showed that strong, optically induced interactions between metamolecules and a light beam with a spatially tailored phase profile can be used to prepare energy ‘hot spots’ of a fraction of a wavelength, which can be created and positioned on the metamaterial landscape. This development offers new opportunities for data storage applications [112, 113] and light harvesting in metamaterial-enhanced radiation detectors [46]. Metasurfaces with strongly interacting metamolecules can exhibit another interesting and useful property of wavevector selectivity. In 2015 we introduced and demonstrated experimental wavevector selective surfaces that are transparent only within a narrow range of light propagation directions, operating effectively as tunnel vision filters [114]. The practical applications of such metasurfaces could include wave-front manipulation, observational instruments and free-space communication in light-scattering environments. One characteristic feature of metamaterials with weakly interacting metamolecules is their insensitivity to disorder and omnidirectional response [115], which is useful for many practical applications.

<sup>5</sup> Dependence of the absorption line width in a gas sample on the sample or beam size is a well-known effect. It has a different nature, which is relevant to the finite interaction time that the atoms flying across the optical beam have in the gas phase.

Another powerful example of metamaterials impacting fundamental physics is the experimental discovery of toroidal dipole response in electrodynamics and the observation of the anapole. The metamaterial platform was crucial for these discoveries. The toroidal dipole is a localized electromagnetic excitation, distinct from the magnetic and electric dipoles. While the electric dipole can be understood as a pair of opposing charges, and the magnetic dipole as a current loop, the toroidal dipole corresponds to currents flowing on the surface of a torus. Toroidal dipoles provide physically significant contributions to the basic characteristics of matter including absorption, dispersion and optical activity. We were interested in toroidal excitation in matter from the very first days of our metamaterial research [116, 117], and in 2004 we hosted the first international workshop on toroidal electrodynamics at Southampton, attended by many earlier researchers on the subject including G N Afanasiev, E V Tkalya, A Ceulemans, M A Martsenyuk, H Schmid and A Dereux. In 2004 I won a very rare and highly sought-after EPSRC adventure grant entitled ‘Supertoroids challenge established laws of physics,’ GR/S48165/01, run with my co-investigators Professor Allan Boardman in Salford University and Professor Tony Bland in Cambridge. Initially, evidence of toroidal response was seen in a chiral metamaterial—an array of wire helix bent into a torus [117]—but the announcement had to wait until 2010, when we clearly saw a toroidal dipole response by detecting a resonant toroidal electromagnetic response in an artificially engineered metamaterial [118]. Observations of toroidal response in terahertz [119], optical plasmonic [120–122] and dielectric metamaterials [123] followed soon together with the developing theory [124].

Another fundamentally important result was the observation of a ‘dynamic anapole’ response in metamaterials. What is a dynamic anapole? Toroidal dipole emitters can be combined with electric dipole emitters, such that the radiated fields interfere destructively. Such a nonradiating configuration is known as a dynamic anapole. Our group reported the observation of the dynamic anapole in a microwave metamaterial in 2013 [125]. The observation of a resonant toroidal response in metamaterials has enabled the systematic study of toroidal electrodynamics [126]. Despite the recent stream of experimental and theoretical works, however, the field is still in its infancy, with many questions to be resolved and applications to be explored. Toroidal resonances in natural media have not yet been observed, and the spectroscopy of toroidal resonances is still being developed [127]. While a strong toroidal contribution is expected only from large toroidal ‘molecules’ (comparable with the wavelength of light), this contribution could be profound [128]. Toroidal resonances can destructively interfere with other modes of excitations in the materials, providing a new mechanism of induced transparency (slow light), and scattering suppression that can be used in narrow-band filters and for dispersion control. Novel laser designs have been investigated where resonant arrays of toroidal metamolecules are used as a gain medium [129]. Finally, toroidal electromagnetic excitations



**Figure 3.** Key metamaterials researchers at the Centre for Photonic Metamaterials of the Optoelectronics Research Centre, University of Southampton in 2016 (from left to right): Edward Rogers, Kevin MacDonald, Nikolay Zheludev, Nikitas Papisimakis, Eric Plum and Vassili Fedotov.

propagating in free space represent an exciting new opportunity for energy and information transfer [130, 131].

### Developing technology and going international

From 2012, we started a very fruitful engagement with Nanyang Technological University (NTU) in Singapore. After winning a very substantial grant from the Ministry of Education there, I was appointed director of the Centre for Disruptive Photonic Technologies (CDPT) at NTU (figure 4) [132]. The centre was built from scratch and is now a two-story complex of research labs and cleanrooms. It is vigorously establishing a reputation as a prominent centre for research and collaboration in south-east Asia, and is one of the main research establishments in nanophotonics and metamaterials in the world. According to the Web of Science, since 2012 NTU has been propelled by CDPT research to the position of leading university in the world in the number of publications on metamaterials (Web of Science search on the topic of ‘metamaterial or metamaterials’ for years 2010–2016). Professors Zexiang Shen, Cesare Soci, David Wilkowski, Hongjin Fan, Sun Handong, Ping Perry Shum, Yidong Chon, Baile Zhang, Ranjan Singh and Weibo Gao joined the centre’s research on metamaterials and have ensured its success. Zexiang Shen and Cesare Soci were appointed co-director and deputy director of the CDPT, and Dr Giorgio Adamo moved from Southampton to NTU to become the research manager of the centre. Other groups at NTU, notably that of Professors Ai Qun Liu and Dao Hua Zhang, also run strong research programmes on metamaterials and collaborate with CDPT. At CDPT we also work closely with our collaborators, and experts in metamaterials at A\*STAR institutions, Professor Boris Lukiyanchuk and Dr Jing Hua Teng, who are both adjunct professors at NTU. Professor Harald Giessen and Federico Capasso have also become NTU visiting professors affiliated with CDPT.

At the CDPT, jointly with the group of Professor Ai Qun Liu, we have worked on MEMS metamaterials driven by comb actuators [133, 134]. We also developed a planar microwave metamaterial in which the resonant properties of every individual metamolecule can be continuously

controlled at will (a random access reconfigurable metamaterial) [135]. To achieve this, we created an array of cavities that can be filled with liquid metal in a controllable fashion, using microfluidic technology and a multiplexer of pneumatic valves. A microfluidic network, addressing every individual element of the array, provides the mechanism to dynamically change the filling factor of the resonators—and thus their resonant electromagnetic properties—continuously and with random access. Such individually addressable cavities of sub-wavelength size form a metamaterial array of subwavelength resonators in which the modulation of the transmitted wavefront is accomplished, with negligible Joule losses, by changing the spatial phase gradient of the array.

Transformation optics—the science of bending light trajectories by filling the space with metamaterials—has always fascinated me, but we never worked on this field in Southampton. However, at NTU we explored some novel ideas on how to build transformation optics and cloaking devices for the optical part of the spectrum. In 2012, in collaboration with Professor Ai Qun Liu, we analysed liquid flowing in an optofluidic waveguide as a new type of controllable medium for transformation optics [136]. We showed that a laminar liquid flow in an optofluidic channel exhibits spatially variable dielectric properties, which support novel wave-focusing and interference phenomena, and which are distinctively different from the discrete diffraction observed in solid waveguide arrays. In 2013, working with Professor Baile Zhang and collaborators in China, we proposed a simplified version of Pendry’s cloak by abolishing the requirement for phase preservation, as it is irrelevant for observation using incoherent natural light with human eyes, which are phase and polarization insensitive. This allows a cloak to be designed on large scales using commonly available materials. We have successfully demonstrated the full optical spectrum cloaking of living creatures, including a cat and a fish, from the human eye [137].

At NTU, we are exploring the use of new materials for metamaterials, such as topological insulators [138, 139] and perovskites [140], as well as studying the quantum effects of light interaction with metamaterials. In 2015 we demonstrated a quantum metamaterial based on the hybridization of plasmonic landscapes with atomic gas [141]. We show that the hyperfine sub-Doppler spectrum of the  $62s_{1/2}-62P_{3/2}$  resonance transition at 852 nm is strongly affected by coupling to the plasmonic resonance of the nanostructure. Fine tuning the dispersion and positions of the atomic lines in the nearfield of plasmonic metamaterials could have uses and implications for atom-based metrology, sensing and the development of atom-on-a-chip devices.

In October 2014, CDPT became a part of The Photonics Institute (TPI), a new institute at NTU coordinated with the ORC at the University of Southampton with research activities spanning the fundamental aspects of nanophotonics, to fibre, telecom sensor and LED technologies. I became the co-director of TPI jointly with Professor Sir David Payne and Professor Swee Chuan Tjin.

Initial funding provided to CDPT by the Ministry of Education in 2012, the research grant on the topic of



**Figure 4.** The opening event for the Center for Disruptive Photonic Technologies at Nanyang Technological University, Singapore, December 2012. The event was attended by many prominent members of the international metamaterial and photonic research community, including Professors David Smith (Duke), Din Ping Tsai (Taiwan), Harald Giessen (Stuttgart), Willi Padilla (Duke), Nader Engheta (UPenn), Anatoly Zayats (Kings), Benjamin Eggleton (Sydney), Michal Lipson (Columbia), John Dudley (Franche-Comté) and Vlad Yakovlev (A&M Texas). The University of Southampton group included Professors Sir David Payne, Dan Hewak, Graham Reed, Micahel Zervas and Gilberto Brambilla. NTU was represented by CDPT Director Professor Nikolay Zheludev, CDPT principle and affiliated investigators Professors Zexiang Shen, Cesare Soci, Baile Zhang, Hongjin Fan, Qijie Wang, David Wilkowski, CDPT research manager Dr Giorgio Adamo and NTU senior management, including the Provost Professor Freddy Boey and the President Professor Bertil Andersson, among other guests. The chief editor of Nature Photonics, Dr Rachel Won, was also present and wrote a feature article in Nature Photonics on the event.

‘disruptive photonic technologies’, MOE2011-T3-1-005, will come to an end in early 2018. However, in 2017 we were able to secure further generous support from the Ministry of Education to continue our work on the topic of ‘quantum and topological nanophotonics’, MOE2016-T3-1-006, the programme in which metamaterials will play an important role. Furthermore, at the end of 2016 we entered the A\*STAR Singapore quantum technologies programme, which looks for applications of metamaterials in quantum devices.

At the University of Southampton 2015 was very significant to us. I was awarded the Thomas Young medal (IOP) for ‘Global leadership and pioneering, seminal work in optical metamaterials and nanophotonics.’ We received a new six-year EPSRC programme grant on the ‘Physics and technology of photonic metadevices and metasystems,’ grant number EP/M009122/1, in which I was appointed principle investigator. At the core of this programme were metamaterials with ‘optical properties on demand’ to facilitate the development of new applications in telecommunication technologies. We have also become a part of the Southampton-centred EPSRC ‘National hub in high value photonic manufacturing,’ EP/N00762X/1 which aims to develop manufacturing processes for advanced photonic technologies, in particular, metamaterials. Professors David Richardson, Otto Muskens, Brian Hayden, assistant Professor Eric Plum and Dr Kevin MacDonald (research manager of the new programme) from Southampton, and Professor Danielle Faccio, a quantum physicist from Harriot Watt University, joined us as

co-investigators. We work closely with other members of the Southampton community, most notably with the theory group of Professor Janne Ruostekoski (School of Mathematics), Dr Xu Fang (ECS), Dr Vassili Fedotov, Assistant Professor Nikitas Papisimakis and Professor-consultant Will Stewart, former chief scientist at Marconi and an early pioneer of metamaterials. Since the Centre for Photonic Metamaterials was established, the Southampton group has become the most cited metamaterials research group in the EU (Web of Science search on the topic of ‘metamaterial or metamaterials’ for the years 2010–2016).

I shall also say that while metamaterials were the main topic of our research from about 2001, we conducted a substantial parallel programme on nanophotonics, which clearly impacted and benefited our work on metamaterials, and in many instances interlinked with it. Above, I gave examples of how our earlier work on nanophotonic switching via structural transformations and active plasmonics (the term which we coined in 2004 [142]) resulted in demonstrations of phase-change memory metamaterials, while the study of electron-beam interactions with nanostructures resulted in the demonstration of frequency, wavelength and beam profile tunable light generation on metasurfaces pumped by free-electron injection. Similarly, our earlier studies of the optical nonlinearities of metals [38–40] and all-optical ultrafast control of plasmon-polaritons [143] have resulted in the development of nonlinear plasmonic metamaterials with strong ultrafast switching [41, 42].

Another interesting and arguably even more important metamaterials-related story is our work on super-resolution imaging. For centuries, improvements in imaging have been drivers of scientific discoveries and technologies, and the metamaterials community was and still is very active in this field—most notably through the massive research of leading groups in the super-resolution negative index Veselago–Pendry superlens and Narimanov–Engheta hyperlens. We stayed away from this mainstream research activity, but developed our own super-resolution approach based on using super-oscillations of light’s field. This came about as the evolution of our 2006 work on light focusing and imaging using nanohole arrays, which are essentially metamaterial lenses [144–146]. Not only has this work evolved into the introduction and development of powerful label-free super-oscillatory imaging, beating the diffraction limit of resolution, it has also pioneered the development of focusing metasurfaces—in particular hole and slit arrays—which have now become a popular topic of research, ten years later.

The development of super-oscillatory imaging technology started in 2006, when Michael Berry and Sandu Popescu at Bristol described [147], and our group in Southampton in 2007 experimentally observed [144] the phenomenon of optical superoscillations. In 2007 we realized the potential of this phenomenon for imaging [145, 146], and in 2009 developed an instructive algorithm for designing super-oscillatory masks [148]. A super-oscillatory imaging device exploits the nontrivial fact that the accurately tailored interference of coherent light waves can form foci that are smaller than those allowed by the Abbe–Rayleigh diffraction limit [149]. In fact there are no fundamental limits on how fine super-oscillatory foci and imaging resolution can be. The

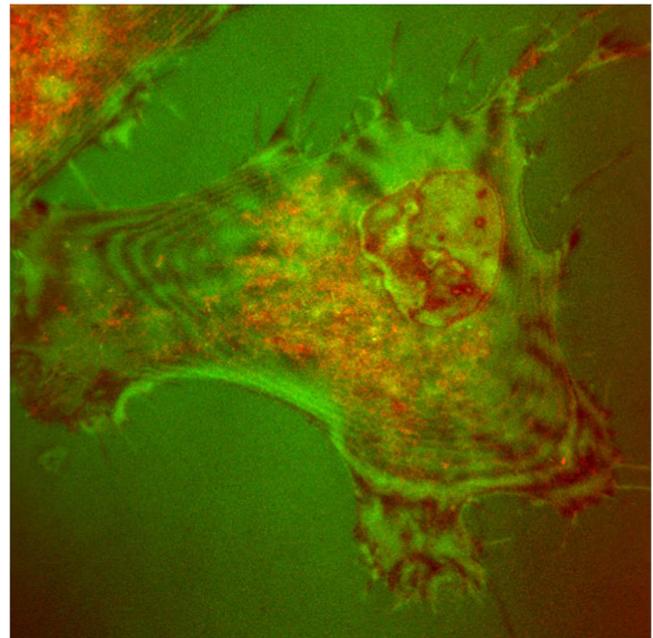
imaging potential of this discovery was rapidly recognized by EPSRC in 2008, when they generously supported the development of super-oscillatory imaging by awarding me a multi-million Basic Technology Research Programme, ‘NANOSCOPE: looking inside a living cell with nanoscale resolution’, EP/F040644/1, which was developed in close collaboration with the groups of Professor Kishan Dholakia from St Andrews, Dr Yifang Chen from the Rutherford Appleton Laboratory, Dr Mark Denis from Bristol and Professor John Chad, a biologist from the University of Southampton.

A practical NANOSCOPE imaging apparatus with a resolution of about one sixth of the wavelength has been demonstrated [150], and a wide range of nanotechnology-enabled super-oscillatory lenses have been developed for applications ranging from imaging to data storage and nanofabrication with light [151–156]. This includes the deployment of super-oscillatory microscopy for bio-imaging [157] and the development of specialized lenses for the visible and infrared spectral bands made from dielectrics and metals, lenses manufactured on fibre tips and silicon wafers, and achromatic as well as apochromatic lenses [158] operating down to single photon level [159]. Dr Edward Rogers, who was initially appointed as a researcher on the Basic Technology Research Programme in 2009 and became a Senior Research Fellow with the Institute for Life Sciences at the University of Southampton and Dr Guanghui Yuan, appointed at the Center for Disruptive Photonic Technologies at NTU Singapore in 2012, have become major contributors to the development of super-oscillatory imaging and focusing technologies.

As we know from recent history, in 2014, Stefan Hell, Eric Betzig and William Moerner received a Nobel Prize for the development of new microscopies that can look into cells at features much smaller than a micron. These techniques require labelling, i.e. the deliberate marking of living cells with dyes and reporters. In the words of the Nobel laureate Eric Betzig, the next revolution in imaging will be in the development of label-free imaging that goes beyond the micron barrier. Indeed, freedom from labels would open up incredible new opportunities for nanotechnology (e.g. lithography), noninvasive and nontoxic biomedical imaging, data storage and precision fabrication with light. Super-oscillatory imaging is a label-free technology with the potential to address all these opportunities. In recent years, the collective effort of optical scientists and biologists at Southampton has advanced to a state in which we can now image the fine details of living cells in real time [157], see figure 5.

## On the future

Where is the metamaterial community heading now and what else is left to do in metamaterials? There is plenty! Let me list just a few directions: random access two- and three-dimensional metamaterials for optical components on demand, reconfigurable photonic circuits, dynamic 3D holographic displays and cognitive metasystems; fast, low-energy all-optical memory components for telecom and displays; large



**Figure 5.** Super-oscillatory unlabelled reflection imaging of MG63 human cells (green) with a conventional image of labelled mitochondria (red) for context (credit to E T F Rogers, S Quraishe, T A Newman, J E Chad, Nikolay I Zheludev and P J S Smith).

area reconfigurable metasurfaces for dynamic paint, self-adaptive camouflage, automotive and fashion industries; high-quality resonance ( $Q > 10\,000$ ) large photonic arrays for switching, lasing, sensing, quantum simulations and quantum computing applications. The high-throughput fabrication of photonic metamaterials remains a major challenge, and we could look at molecular manufacturing using kinematic self-replicating machines, diamond mechanosynthesis, DNA scaffolding etc. Metamaterials for UV, soft x-ray and the matter waves of ultralow temperature molecular/atomic beams are still waiting for their time to come!

In conclusion, I would like to answer an important question. With all the remarkable success and impact of the metamaterial research programmes internationally, the emergence of a mighty and highly skilled research community, and substantial funding across the globe, why do we not see the massive deployment of metamaterials yet? Many ask this question, and there are several reasons one would need to take into consideration when answering it. First, we need to admit that the field was partially discredited by overstated promises and hype in the early days of the discipline, and we all have to take responsibility for this. This has made the industry over-cautions in embracing metamaterials. Second, for a few years, due to enormous popular interest in the subject, metamaterials were seen as only relevant to superlenses and cloaking-device applications, which limited them in the eyes of the general public and industrial developers, obscuring their real practical impact on other applications. This perception is rapidly changing, but it will take time for the industry to appreciate the true potential of metamaterials across a wide range of technologies. Third, metamaterials are not a technology as such, but are technology enablers, and thus their proliferation

faces a number of challenges. For example, greater education of end-users is required in many application fields; additionally, the development of high-throughput, cost-effective fabrication processes is needed. Most importantly, the development of integration approaches, such as metamaterial + fibre technology, metamaterial + silicon photonic technology, and metamaterial-enabled photovoltaics is crucial. And finally, more time is needed: from the discovery of the light emitting diode in 1925 and LED-based optical telecommunication in 1927 [160], more than 60 years was required for the massive deployment of the optical telecommunications seen currently. As of today, the field of metamaterials is only 16 years old.

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Papasimakis N, Perino M, Petropoulos P, Plum E, Pochon S, Popov S V, Potts A, Prosvirnin S L, Quraishie S, Rao S M, Raybould T A, Ren M X, Richardson D J, Rockstuhl C, Rogacheva A V, Roger T, Rogers E T F, Rose M, Roy T, Ruostekoski J, Samson Z L, Savinov V, Savo S, Schultz S, Schwanecke A S, Shatwell I R, Shen L, Shen Z X, Shi J H, Singh R, Slobodchikov E V, Smith D R, Smith P J S, So J K, Soares B F, Song Q H, Soukoulis C, Stevens G, Stockman M I, Sulaev A, Svirko Y P, Tanaka K, Tao J R, Teng J H, Tsai D P, Tsai W Y, Tseng M L, Tsiatmas A, Uchino T, Valente J, Vezzoli S, Vier D C, Wallauer J, Walther M, Wang C M, Wang H P, Wang L, Wang Q, Wang S, Waters R F, Wilkowski D, Wu P C, Xu J J, Xu N, Yan L B, Yang Y, Youngs I, Yuan G H, Zhang B L, Zhang J B, Zhang J F, Zhang J, Zhang W, Zhang X M, Zhao X Z, Zheng B, Zhou J, Zhu W M.

## References

- [1] Feynman R P 1992 Plenty of room at the bottom *J. Microelectromech. Syst.* **1** 60–6
- [2] Zheludev N I 2010 The road ahead for metamaterials *Science* **328** 582–3
- [3] Zheludev N I 2015 Obtaining optical properties on demand *Science* **348** 973–4
- [4] Svirko Y P and Zheludev N I 1998 *Polarization of Light in Nonlinear Optics* (Chichester: Wiley)
- [5] Bennett P J, Dhanjal S, Petropoulos P, Richardson D J, Zheludev N I and Emelyanov V I 1998 A photonic switch based on a gigantic, reversible optical nonlinearity of liquefying gallium *Appl. Phys. Lett.* **73** 1787–9
- [6] Smith D R, Padilla W J, Vier D C, Nemat-Nasser S C and Schultz S 2000 Composite medium with simultaneously negative permeability and permittivity *Phys. Rev. Lett.* **84** 4184–7
- [7] Svirko Y, Zheludev N and Osipov M 2001 Layered chiral metallic microstructures with inductive coupling *Appl. Phys. Lett.* **78** 498–500
- [8] Shalaev V M *et al* 2005 Negative index of refraction in optical metamaterials *Opt. Lett.* **30** 3356–8
- [9] Papakostas A, Potts A, Bagnall D, Prosvirnin S, Coles H and Zheludev N 2003 Optical manifestations of planar chirality *Phys. Rev. Lett.* **90** 107404
- [10] Yu N F *et al* 2011 Light propagation with phase discontinuities: generalized laws of reflection and refraction *Science* **334** 333–7
- [11] Fedotov V, Mladyonov P, Prosvirnin S, Rogacheva A, Chen Y and Zheludev N 2006 Asymmetric propagation of electromagnetic waves through a planar chiral structure *Phys. Rev. Lett.* **97** 167401
- [12] Fedotov V A, Schwanecke A S, Zheludev N I, Khardikov V V and Prosvirnin S L 2007 Asymmetric transmission of light and enantiomerically sensitive plasmon resonance in planar chiral nanostructures *Nano Lett.* **7** 1996–9
- [13] Schwanecke A S, Fedotov V A, Khardikov V V, Prosvirnin S L, Chen Y and Zheludev N I 2008 Nanostructured metal film with asymmetric optical transmission *Nano Lett.* **8** 2940–3
- [14] Singh R *et al* 2009 Terahertz metamaterial with asymmetric transmission *Phys. Rev. B* **80** 153104
- [15] Fedotov V A, Mladyonov P L, Prosvirnin S L and Zheludev N I 2005 Planar electromagnetic metamaterial with a fish scale structure *Phys. Rev. E* **72** 056613

- [16] Fedotov V A, Rogacheva A V, Zheludev N I, Mladyonov P L and Prosvirnin S L 2006 Mirror that does not change the phase of reflected waves *Appl. Phys. Lett.* **88** 091119
- [17] Schwanecke A S, Fedotov V A, Khardikov V V, Prosvirnin S L, Chen Y and Zheludev N I 2007 Optical magnetic mirrors *J. Opt. A: Pure Appl. Opt.* **9** L1–2
- [18] Alu A and Engheta N 2005 Achieving transparency with plasmonic and metamaterial coatings *Phys. Rev. E* **72** 016623
- [19] Chen Y F, Tao J R, Zhao X Z, Cui Z, Schwanecke A S and Zheludev N I 2005 Nanoimprint lithography for planar chiral photonic meta-materials *Microelectron. Eng.* **78–79** 612–7
- [20] Rogacheva A V, Fedotov V A, Schwanecke A S and Zheludev N I 2006 Giant gyrotropy due to electromagnetic-field coupling in a bilayered chiral structure *Phys. Rev. Lett.* **97** 177401
- [21] Plum E *et al* 2009 Metamaterial with negative index due to chirality *Phys. Rev. B* **79** 035407
- [22] Plum E, Fedotov V A, Schwanecke A S, Zheludev N I and Chen Y 2007 Giant optical gyrotropy due to electromagnetic coupling *Appl. Phys. Lett.* **90** 223113
- [23] Plum E, Liu X X, Fedotov V A, Chen Y, Tsai D P and Zheludev N I 2009 Metamaterials: optical activity without chirality *Phys. Rev. Lett.* **102** 113902
- [24] Plum E, Fedotov V A and Zheludev N I 2016 Specular optical activity of achiral metasurfaces *Appl. Phys. Lett.* **108** 141905
- [25] Plum E and Zheludev N I 2015 Chiral mirrors *Appl. Phys. Lett.* **106** 221901
- [26] Plum E, Fedotov V A and Zheludev N I 2009 Extrinsic electromagnetic chirality in metamaterials *J. Opt. A: Pure Appl. Opt.* **11** 074009
- [27] Plum E, Fedotov V A and Zheludev N I 2011 Asymmetric transmission: a generic property of two-dimensional periodic patterns *J. Opt.* **13** 024006
- [28] Fedotov V A, Rose M, Prosvirnin S L, Papasimakis N and Zheludev N I 2007 Sharp trapped-mode resonances in planar metamaterials with a broken structural symmetry *Phys. Rev. Lett.* **99** 147401
- [29] Luk'yanchuk B *et al* 2010 The Fano resonance in plasmonic nanostructures and metamaterials *Nat. Mater.* **9** 707–15
- [30] Papasimakis N, Fedotov V A, Zheludev N I and Prosvirnin S L 2008 Metamaterial analog of electromagnetically induced transparency *Phys. Rev. Lett.* **101** 253903
- [31] Papasimakis N and Zheludev N I 2009 Metamaterial-induced transparency *Opt. Photonics News* **20** 22–7
- [32] Stockman M I 2013 Lasing spaser in two-dimensional plasmonic crystals *NPG Asia Mater.* **5** e71
- [33] Tanaka K, Plum E, Ou J Y, Uchino T and Zheludev N I 2010 Multifold enhancement of quantum dot luminescence in plasmonic metamaterials *Phys. Rev. Lett.* **105** 227403
- [34] Graydon O 2017 A question of topology *Nat. Photon.* **11** 146–7
- [35] Nikolaenko A E *et al* 2010 Carbon nanotubes in a photonic metamaterial *Phys. Rev. Lett.* **104** 153902
- [36] Papasimakis N *et al* 2010 Graphene in a photonic metamaterial *Opt. Express* **18** 8353–9
- [37] Nikolaenko A E *et al* 2012 Nonlinear graphene metamaterial *Appl. Phys. Lett.* **100** 181109
- [38] Dhanjal S, Popov S V, Shatwell I R, Svirko Y P, Zheludev N I and Gusev V E 1997 Femtosecond optical nonlinearity of metallic indium across the solid-liquid transition *Opt. Lett.* **22** 1879–81
- [39] Bennett P J, Dhanjal S, Svirko Y P and Zheludev N I 1998 Cubic optical nonlinearities of metals in the vicinity of the melting point *J. Mod. Opt.* **45** 1009–18
- [40] Zheludev N I *et al* 1995 Cubic optical nonlinearity of free-electrons in bulk gold *Opt. Lett.* **20** 1368–70
- [41] Ren M X *et al* 2011 Nanostructured plasmonic medium for terahertz bandwidth all-optical switching *Adv. Mater.* **23** 5540–4
- [42] Ren M X, Plum E, Xu J J and Zheludev N I 2012 Giant nonlinear optical activity in a plasmonic metamaterial *Nat. Commun.* **3** 833
- [43] Tsiatmas A *et al* 2010 Superconducting plasmonics and extraordinary transmission *Appl. Phys. Lett.* **97** 111106
- [44] Tsiatmas A, Fedotov V A, de Abajo F J G and Zheludev N I 2012 Low-loss terahertz superconducting plasmonics *New J. Phys.* **14** 115006
- [45] Fedotov V A *et al* 2010 Temperature control of Fano resonances and transmission in superconducting metamaterials *Opt. Express* **18** 9015–9
- [46] Savinov V, Fedotov V A, de Groot P A and Zheludev N I 2013 Radiation-harvesting resonant superconducting sub-THz metamaterial bolometer *Supercond. Sci. Technol.* **26** 084001
- [47] Savinov V, Fedotov V A, Anlage S M, de Groot P A J and Zheludev N I 2012 Modulating sub-THz radiation with current in superconducting metamaterial *Phys. Rev. Lett.* **109** 243904
- [48] Savinov V, Tsiatmas A, Buckingham A R, Fedotov V A, de Groot P A J and Zheludev N I 2012 Flux exclusion superconducting quantum metamaterial: towards quantum-level switching *Sci. Rep.* **2** 450
- [49] Savinov V, Delfanazari K, Fedotov V A and Zheludev N I 2016 Giant nonlinearity in a superconducting sub-terahertz metamaterial *Appl. Phys. Lett.* **108** 101107
- [50] Zheludev N 2007 All change, please *Nat. Photon.* **1** 551–3
- [51] Gupta M *et al* 2016 Sharp toroidal resonances in planar terahertz metasurfaces *Adv. Mater.* **28** 8206–11
- [52] Zheludev N I 2002 Nonlinear optics on the nanoscale *Contemp. Phys.* **43** 365–77
- [53] Zheludev N I 2006 Single nanoparticle as photonic switch and optical memory element *J. Opt. A: Pure Appl. Opt.* **8** S1–8
- [54] Albanis V *et al* 2001 Nanosecond dynamics of a gallium mirror's light-induced reflectivity change *Phys. Rev. B* **63** 165207
- [55] Soares B F, MacDonald K F, Fedotov V A and Zheludev N I 2005 Light-induced switching between structural forms with different optical properties in a single gallium nanoparticle *Nano Lett.* **5** 2104–7
- [56] Soares B F, Jonsson F and Zheludev N I 2007 All-optical phase-change memory in a single gallium nanoparticle *Phys. Rev. Lett.* **98** 153905
- [57] MacDonald K F, Fedotov V A and Zheludev N I 2003 Optical nonlinearity resulting from a light-induced structural transition in gallium nanoparticles *Appl. Phys. Lett.* **82** 1087–9
- [58] MacDonald K F *et al* 2004 Controlling the coexistence of structural phases and the optical properties of gallium nanoparticles with optical excitation *Europhys. Lett.* **67** 614–9
- [59] Krasavin A V, MacDonald K F, Schwanecke A S and Zheludev N I 2006 Gallium/aluminum nanocomposite material for nonlinear optics and nonlinear plasmonics *Appl. Phys. Lett.* **89** 031118
- [60] Waters R F, Hobson P A, MacDonald K F and Zheludev N I 2015 Optically switchable photonic metasurfaces *Appl. Phys. Lett.* **107** 081102
- [61] Samson Z L *et al* 2010 Metamaterial electro-optic switch of nanoscale thickness *Appl. Phys. Lett.* **96** 143105
- [62] Gholipour B, Zhang J, MacDonald K F, Hewak D W and Zheludev N I 2013 An all-optical, non-volatile, bidirectional, phase-change meta-switch *Adv. Mater.* **25** 1–5

- [63] Wang Q *et al* 2016 Optically reconfigurable metasurfaces and photonic devices based on phase change materials *Nat. Photon.* **10** 60–5
- [64] Wang Q, Yuan G H, Kiang K S, Sun K, Gholipour B, Rogers E T F, Huang K, Ang S S, Zheludev N I and Teng J H 2017 Reconfigurable phase-change photomask for grayscale photolithography *Appl. Phys. Lett.* **110** 201110
- [65] Zheludev N I and Plum E 2016 Reconfigurable nanomechanical photonic metamaterials *Nat. Nanotechnol.* **11** 16–22
- [66] Ou J Y, Plum E, Jiang L and Zheludev N I 2011 Reconfigurable photonic metamaterials *Nano Lett.* **11** 2142–4
- [67] Valente J, Plum E, Youngs I J and Zheludev N I 2016 Nano- and micro-auxetic plasmonic materials *Adv. Mater.* **28** 5176–80
- [68] Ou J Y, Plum E, Zhang J F and Zheludev N I 2013 An electromechanically reconfigurable plasmonic metamaterial operating in the near-infrared *Nat. Nanotechnol.* **8** 252–5
- [69] Valente J, Ou J Y, Plum E, Youngs I J and Zheludev N I 2015 A magneto-electro-optical effect in a plasmonic nanowire material *Nat. Commun.* **6** 7021
- [70] Ou J Y, Plum E, Zhang J F and Zheludev N I 2016 Giant nonlinearity of an optically reconfigurable plasmonic metamaterial *Adv. Mater.* **28** 729–33
- [71] Karvounis A, Ou J Y, Wu W P, MacDonald K F and Zheludev N I 2015 Nano-optomechanical nonlinear dielectric metamaterials *Appl. Phys. Lett.* **107** 191110
- [72] Cencillo-Abad P, Ou J Y, Plum E, Valente J and Zheludev N I 2016 Random access actuation of nanowire grid metamaterial *Nanotechnology* **27** 485206
- [73] Cencillo-Abad P, Plum E, Rogers E T F and Zheludev N I 2016 Spatial optical phase-modulating metadvice with subwavelength pixelation *Opt. Express* **24** 18790–8
- [74] Cencillo-Abad P, Zheludev N I and Plum E 2016 Metadvice for intensity modulation with sub-wavelength spatial resolution *Sci. Rep.* **6** 37109
- [75] Zhang J F, MacDonald K F and Zheludev N I 2013 Nonlinear dielectric optomechanical metamaterials *Light Sci. Appl.* **2** e96
- [76] Zhang J, MacDonald K F and Zheludev N I 2012 Controlling light-with-light without nonlinearity *Light Sci. Appl.* **1** e18
- [77] Shi J H, Fang X, Rogers E T F, Plum E, MacDonald K F and Zheludev N I 2014 Coherent control of Snell's law at metasurfaces *Opt. Express* **22** 21051–60
- [78] Mousavi S A, Plum E, Shi J H and Zheludev N I 2014 Coherent control of birefringence and optical activity *Appl. Phys. Lett.* **105** 011906
- [79] Rao S M, Lyons A, Roger T, Clerici M, Zheludev N I and Faccio D 2015 Geometries for the coherent control of four-wave mixing in graphene multilayers *Sci. Rep.* **5** 15399
- [80] Fang X, MacDonald K F and Zheludev N I 2015 Controlling light with light using coherent metadvice: all-optical transistor, summator and inverter *Light Sci. Appl.* **4** e292
- [81] Roger T *et al* 2015 Coherent perfect absorption in deeply subwavelength films in the single-photon regime *Nat. Commun.* **6** 7031
- [82] Fang X, Tseng M L, Ou J Y, MacDonald K F, Tsai D P and Zheludev N I 2014 Ultrafast all-optical switching via coherent modulation of metamaterial absorption *Appl. Phys. Lett.* **104** 141102
- [83] Nalla V *et al* 2015 *Metamaterials 2015 (Oxford, 7–12 September 2015)*
- [84] Nalla V, Karvounis A, Valente J, Sun H D and Zheludev N I 2017 Ultrafast regimes of coherent absorption in plasmonic and dielectric diamond-based metasurfaces *ICMAT2017 (Singapore, 18–23 June 2017)*
- [85] Fang X, Tseng M L, Tsai D P and Zheludev N I 2016 Coherent excitation-selective spectroscopy of multipole resonances *Phys. Rev. Appl.* **5** 014010
- [86] Fang X, MacDonald K F, Plum E and Zheludev N I 2016 Coherent control of light–matter interactions in polarization standing waves *Sci. Rep.* **6** 31141
- [87] Tseng M L *et al* 2017 Coherent selection of invisible high-order electromagnetic excitations *Sci. Rep.* **7** 44488
- [88] Papaioannou M, Plum E, Valente J, Rogers E T F and Zheludev N I 2016 Two-dimensional control of light with light on metasurfaces *Light Sci. Appl.* **5** e16070
- [89] Papaioannou M, Plum E and Zheludev N I 2017 All-optical pattern recognition and image processing on a metamaterial beam splitter *ACS Photonics* **4** 217–22
- [90] Papaioannou M, Plum E, Valente J, Rogers E T F and Zheludev N I 2016 All-optical multichannel logic based on coherent perfect absorption in a plasmonic metamaterial *APL Photonics* **1** 090801
- [91] Pochon S, MacDonald K F, Knize R J and Zheludev N I 2004 Phase coexistence in gallium nanoparticles controlled by electron excitation *Phys. Rev. Lett.* **92** 145702
- [92] Bashevoy M V, Jonsson F, Krasavin A V, Zheludev N I, Chen Y and Stockman M I 2006 Generation of traveling surface plasmon waves by free-electron impact *Nano Lett.* **6** 1113–5
- [93] So J K, de Abajo F J G, MacDonald K F and Zheludev N I 2015 Amplification of the evanescent field of free electrons *ACS Photonics* **2** 1236–40
- [94] Adamo G *et al* 2009 Light well: a tunable free-electron light source on a chip *Phys. Rev. Lett.* **103** 113901
- [95] Adamo G *et al* 2012 Electron-beam-driven collective-mode metamaterial light source *Phys. Rev. Lett.* **109** 217401
- [96] Li G, Clarke B P, So J K, MacDonald K F and Zheludev N I 2016 Holographic free-electron light source *Nat. Commun.* **7** 13705
- [97] Zhang J F, Ou J Y, Papisimakis N, Chen Y F, MacDonald K F and Zheludev N I 2011 Continuous metal plasmonic frequency selective surfaces *Opt. Express* **19** 23279–85
- [98] Zhang J, Ou J Y, MacDonald K F and Zheludev N I 2012 Optical response of plasmonic relief meta-surfaces *J. Opt.* **14** 114002
- [99] Fedotov V A, Emel'yanov V I, MacDonald K F and Zheludev N I 2004 Optical properties of closely packed nanoparticle films: spheroids and nanoshells *J. Opt. A: Pure Appl. Opt.* **6** 155–60
- [100] Zhang J F, MacDonald K F and Zheludev N I 2013 Near-infrared trapped mode magnetic resonance in an all-dielectric metamaterial *Opt. Express* **21** 26721–8
- [101] Zhang J F, MacDonald K F and Zheludev N I 2014 Giant optical forces in planar dielectric photonic metamaterials *Opt. Lett.* **39** 4883–6
- [102] Karvounis A, Gholipour B, MacDonald K F and Zheludev N I 2016 All-dielectric phase-change reconfigurable metasurface *Appl. Phys. Lett.* **109** 051103
- [103] Gholipour B *et al* 2017 Organometallic perovskite metasurfaces *Adv. Mater.* **29** 1604268
- [104] Jenkins S D and Ruostekoski J 2013 Metamaterial transparency induced by cooperative electromagnetic interactions *Phys. Rev. Lett.* **111** 147401
- [105] Jenkins S D and Ruostekoski J 2012 Resonance linewidth and inhomogeneous broadening in a metamaterial array *Phys. Rev. B* **86** 205128
- [106] Jenkins S D and Ruostekoski J 2012 Cooperative resonance linewidth narrowing in a planar metamaterial *New J. Phys.* **14** 103003
- [107] Jenkins S D and Ruostekoski J 2012 Controlled manipulation of light by cooperative response of atoms in an optical lattice *Phys. Rev. A* **86** 031602

- [108] Jenkins S D and Ruostekoski J 2012 Theoretical formalism for collective electromagnetic response of discrete metamaterial systems *Phys. Rev. B* **86** 205128
- [109] Fedotov V A *et al* 2010 Spectral collapse in ensembles of metamolecules *Phys. Rev. Lett.* **104** 223901
- [110] Papasimakis N, Fedotov V A, Fu Y H, Tsai D P and Zheludev N I 2009 Coherent and incoherent metamaterials and order-disorder transitions *Phys. Rev. B* **80** 041102
- [111] Savo S, Papasimakis N and Zheludev N I 2012 Localization of electromagnetic fields in disordered metamaterials *Phys. Rev. B* **85** 121104
- [112] Kao T S, Huang F M, Chen Y, Rogers E T F and Zheludev N I 2010 Metamaterial as a controllable template for nanoscale field localization *Appl. Phys. Lett.* **96** 041103
- [113] Kao T S, Jenkins S D, Ruostekoski J and Zheludev N I 2011 Coherent control of nanoscale light localization in metamaterial: creating and positioning isolated subwavelength energy hot spots *Phys. Rev. Lett.* **106** 085501
- [114] Fedotov V A, Wallauer J, Walther M, Perino M, Papasimakis N and Zheludev N I 2015 Wavevector selective metasurfaces and tunnel vision filters *Light Sci. Appl.* **4** e306
- [115] Papasimakis N, Fu Y, Fedotov V, Prosvirnin S, Tsai D and Zheludev N 2009 Metamaterial with polarization and direction insensitive resonant transmission response mimicking electromagnetically induced transparency *Appl. Phys. Lett.* **94** 211902
- [116] Marinov K, Boardman A D, Fedotov V A and Zheludev N 2007 Toroidal metamaterial *New J. Phys.* **9** 324
- [117] Papasimakis N, Fedotov V A, Marinov K and Zheludev N I 2009 Gyrotropy of a metamolecule: wire on a torus *Phys. Rev. Lett.* **103** 093901
- [118] Kaelberer T, Fedotov V A, Papasimakis N, Tsai D P and Zheludev N I 2010 Toroidal dipolar response in a metamaterial *Science* **330** 1510–2
- [119] Gupta M *et al* 2016 Sharp toroidal resonances in planar terahertz metasurfaces *Adv. Mater.* **28** 8206–11
- [120] Wu P C *et al* 2014 Three-dimensional metamaterials: from split ring resonator to toroidal metamolecule *Proc. SPIE* **9163** 91630M
- [121] Liao C Y *et al* 2015 Optical toroidal response in three-dimensional plasmonic metamaterial *Proc. SPIE* **9547** 954724
- [122] Huang Y W *et al* 2012 Design of plasmonic toroidal metamaterials at optical frequencies *Opt. Express* **20** 1760–8
- [123] Basharin A A *et al* 2015 Dielectric metamaterials with toroidal dipolar response *Phys. Rev. X* **5** 011036
- [124] Watson D W, Jenkins S D, Ruostekoski J, Fedotov V A and Zheludev N I 2016 Toroidal dipole excitations in metamolecules formed by interacting plasmonic nanorods *Phys. Rev. B* **93** 125420
- [125] Fedotov V A, Rogacheva A V, Savinov V, Tsai D P and Zheludev N I 2013 Resonant transparency and non-trivial non-radiating excitations in toroidal metamaterials *Sci. Rep.* **3** 2967
- [126] Papasimakis N, Fedotov V A, Savinov V, Raybould T A and Zheludev N I 2016 Electromagnetic toroidal excitations in matter and free space *Nat. Mater.* **15** 263–71
- [127] Savinov V, Fedotov V A and Zheludev N I 2014 Toroidal dipolar excitation and macroscopic electromagnetic properties of metamaterials *Phys. Rev. B* **89** 205112
- [128] Raybould T A *et al* 2016 Toroidal circular dichroism *Phys. Rev. B* **94** 035119
- [129] Huang Y W, Chen W T, Wu P C, Fedotov V A, Zheludev N I and Tsai D P 2013 Toroidal lasing spaser *Sci. Rep.* **3** 1237
- [130] Raybould T, Fedotov V, Papasimakis N, Youngs I and Zheludev N 2016 Focused electromagnetic doughnut pulses and their interaction with interfaces and nanostructures *Opt. Express* **24** 3150–61
- [131] Raybould T, Papasimakis N, Fedotov V A, Youngs I and Zheludev N I 2017 Generation of flying electromagnetic doughnuts via spatiothermal conversion of transverse electromagnetic pulses *Nanometa 2017 (Seefeld, Austria, 4–7 January 2017)*
- [132] Won R 2013 Research: new centre, new vision *Nat. Photon.* **7** 171
- [133] Zhu W M *et al* 2012 Microelectromechanical Maltese-cross metamaterial with tunable terahertz anisotropy *Nat. Commun.* **3** 1274
- [134] Fu Y H *et al* 2011 A micromachined reconfigurable metamaterial via reconfiguration of asymmetric split-ring resonators *Adv. Funct. Mater.* **21** 3589–94
- [135] Zhu W M *et al* 2015 A flat lens with tunable phase gradient by using random access reconfigurable metamaterial *Adv. Mater.* **27** 4739–43
- [136] Yang Y *et al* 2012 Optofluidic waveguide as a transformation optics device for lightwave bending and manipulation *Nat. Commun.* **3** 651
- [137] Chen H S *et al* 2013 Ray-optics cloaking devices for large objects in incoherent natural light *Nat. Commun.* **4** 2652
- [138] Ou J Y, So J K, Adamo G, Sulaev A, Wang L and Zheludev N I 2014 Ultraviolet and visible range plasmonics in the topological insulator  $\text{Bi}_{1.5}\text{Sb}_{0.5}\text{Te}_{1.8}\text{Se}_{1.2}$  *Nat. Commun.* **5** 5139
- [139] Dubrovkin A M, Adamo G, Yin J, Wang L, Soci C, Wang Q J and Zheludev N I 2017 Visible range plasmonic modes on topological insulator nanostructures *Adv. Opt. Mater.* **5** 1600768
- [140] Gholipour B, Adamo G, Cortecchia D, Krishnamoorthy N S, Birowosuto M D, Zheludev N I and Soci C 2016 Oranometallic Perovskite Metasurfaces *Adv. Mater.* **29** 1604268
- [41] Aljunid S A, Chan E A, Adamo G, Ducloy M, Wilkowski D and Zheludev N I 2016 Atomic response in the near-field of nanostructured plasmonic metamaterial *Nano Lett.* **16** 3137–41
- [142] Krasavin A V and Zheludev N 2004 Active plasmonics: controlling signals in Au/Ga waveguide using nanoscale structural transformations *Appl. Phys. Lett.* **84** 1416–8
- [143] MacDonald K F, Samson Z L, Stockman M I and Zheludev N I 2009 Ultrafast active plasmonics *Nat. Photon.* **3** 55–8
- [144] Huang F M, Zheludev N, Chen Y F and de Abajo F J G 2007 Focusing of light by a nanohole array *Appl. Phys. Lett.* **90** 091119
- [145] Huang F M, Kao T S, Fedotov V A, Chen Y F and Zheludev N I 2008 Nanohole array as a lens *Nano Lett.* **8** 2469–72
- [146] Huang F M, Chen Y, de Abajo F J G and Zheludev N I 2007 Optical super-resolution through super-oscillations *J. Opt. A: Pure Appl. Opt.* **9** S285–8
- [147] Berry M V and Popescu S 2006 Evolution of quantum superoscillations and optical superresolution without evanescent waves *J. Phys. A: Math. Gen.* **39** 6965–77
- [148] Huang F M and Zheludev N I 2009 Super-resolution without evanescent waves *Nano Lett.* **9** 1249–54
- [149] Rogers E T F and Zheludev N I 2013 Optical super-oscillations: sub-wavelength light focusing and super-resolution imaging *J. Opt.* **15** 1–23
- [150] Rogers E T F *et al* 2012 A super-oscillatory lens optical microscope for subwavelength imaging *Nat. Mater.* **11** 432–5
- [151] Rogers E T F, Savo S, Lindberg J, Roy T, Dennis M R and Zheludev N I 2013 Super-oscillatory optical needle *Appl. Phys. Lett.* **102** 031108
- [152] Roy T, Rogers E T F and Zheludev N I 2013 Sub-wavelength focusing meta-lens *Opt. Express* **21** 7577–82

- [153] Roy T, Rogers E T F, Yuan G H and Zheludev N I 2014 Point spread function of the optical needle super-oscillatory lens *Appl. Phys. Lett.* **104** 231109
- [154] Yuan G H, Rogers E T F, Roy T, Adamo G, Shen Z X and Zheludev N I 2014 Planar super-oscillatory lens for sub-diffraction optical needles at violet wavelengths *Sci. Rep.* **4** 6333
- [155] Yuan G H, Rogers E T F, Roy T, Shen Z X and Zheludev N I 2014 Flat super-oscillatory lens for heat-assisted magnetic recording with sub-50 nm resolution *Opt. Express* **22** 6428–37
- [156] Baumgartl J, Kosmeier S, Mazilu M, Rogers E T F, Zheludev N I and Dholakia K 2011 Far field subwavelength focusing using optical eigenmodes *Appl. Phys. Lett.* **98** 181109
- [157] Rogers E T F *et al* 2015 Super-oscillatory imaging of nanoparticle interactions with neurons *Biophys. J.* **108** 479a–79a
- [158] Yuan G H, Rogers E T F and Zheludev N I 2017 Achromatic super-oscillatory lenses with sub-wavelength focusing *Light Sci. Appl.* **6** e17036
- [159] Yuan G H *et al* 2016 Quantum super-oscillation of a single photon *Light Sci. Appl.* **5** e16127
- [160] Zheludev N 2007 The life and times of the LED—a 100-year history *Nat. Photon.* **1** 189–92