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Plasmonics: An Emerging Field Fostered by *Nano Letters*

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ABSTRACT While studies of surface plasmons on metals have been pursued for decades, the more recent appearance of nanoscience has created a revolution in this field with "Plasmonics" emerging as a major area of research. The direct optical excitation of surface plasmons on metallic nanostructures provides numerous ways to control and manipulate light at nanoscale dimensions. This has stimulated the development of novel optical materials, deeper theoretical insight, innovative new devices, and applications with potential for significant technological and societal impact. *Nano Letters* has been instrumental in the emergence of plasmonics, providing its readership with rapid advances in this dynamic field.

KEYWORDS Surface plasmons, plasmonics, nanophotonics, nano-optics, sensing, nanoscale devices

Metallic nanoparticles have long been known for their optical properties. The colorful stained glass windows of medieval Europe exploited these properties long before modern science provided an understanding of their origin. Just over a century ago, classical electromagnetic theory provided the first rigorous explanation of the optical properties of noble metal nanoparticles, explaining the unusually vibrant red color of gold colloid.[1] In the intervening decades, our understanding of surface electromagnetic waves, and the collective excitations of metals known as surface plasmons, has developed and matured for macroscopic metals[2] as well as for small particles.[3] As a fundamental excitation of condensed matter, surface plasmons have been studied extensively using surface science techniques and many-body theory. Surface plasmons were primarily perceived as excitations whose properties are characteristic of specific metals. Interest in this topic was dominated by explanations of plasmon energies and dispersion characteristics relative to the electronic structure of the metal.

Plasmonics, as a discipline that has emerged since the initial issues of *Nano Letters*, is truly emblematic of modern interdisciplinary nanoscience.

With the rise of nanoscience, the realization that surface plasmon properties are also largely controlled by the shape of a metal structure at subwavelength dimensions has resulted in a tremendous increase in interest in plasmon-based phenomena. With the confluence of plasmon science and nanoscience came the understanding that plasmonic properties could be manipulated, essentially designed, by the selection and fabrication of metallic structures of specific geometries. For metal objects with dimensions smaller than the wavelength of light, surface waves can be excited by direct optical excitation, leading to the conception of metallic or "plasmonic" nanostructures as nanoscale optical components. The concurrent, and fortuitous, availability of high-performance computational electromagnetic simulation software, enabling close agreement between observed and predicted properties of metallic structures, has resulted in advanced design, even quantitative capabilities, enabling rapid advances in this field. This design component is unique within nanoscience and has resulted in a broad interest in plasmonics across a range of diverse disciplines from condensed matter physics to computer chip design and biomedical engineering. Plasmonics, as a discipline that has emerged since the initial issues of *Nano Letters*, is truly emblematic of modern interdisciplinary nanoscience, arising at the intersection of multiple fields: condensed matter physics, materials and physical chemistry, and electrical engineering. As one of the first journals to encourage submission from a wide range of fields, *Nano Letters* fostered the development of plasmonics by providing a home for high quality papers on this topic spanning a broad range of disciplines. Applications of plasmonic structures have emerged in areas both within and beyond this traditional scope with particular interest in applications in biology, biotechnology, and biomedicine.

Shape Matters. Since the early days of *Nano Letters*, the chemical synthesis of noble metal nanoparticles of

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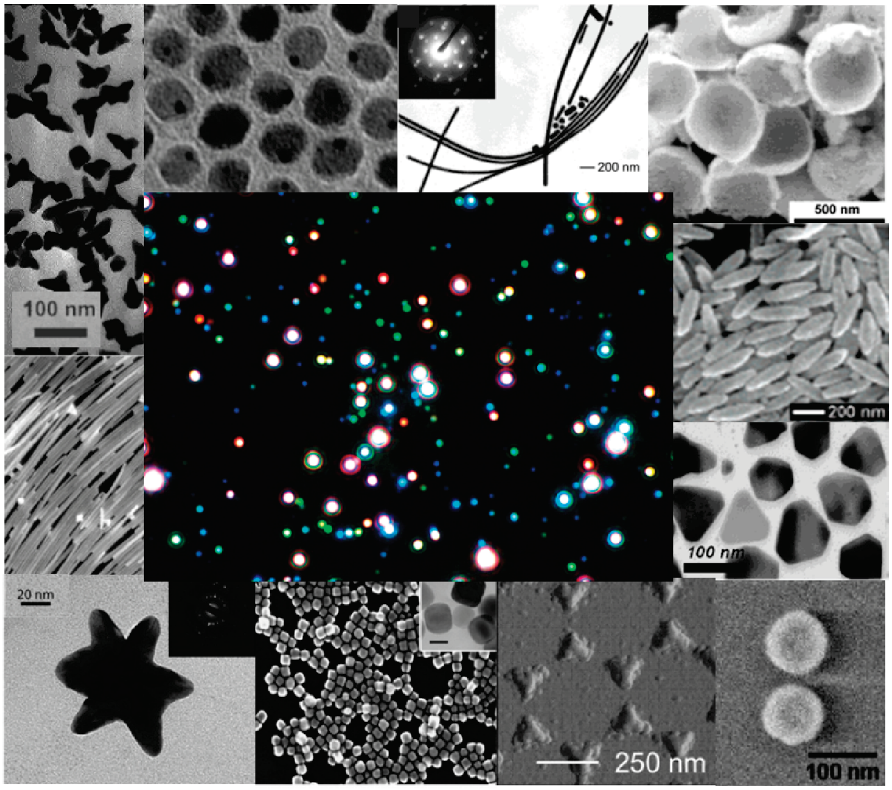

FIGURE 1. Plasmonic nanoparticles and nanostructures appearing in *Nano Letters*. Clockwise from top left: branched Au nanocrystals;[4] dumbbell-like bifunctional Au-Fe3O4 nanoparticles;[5] Ag nanowires directly coated with amorphous silica;[6] metallic half-shells with submicrometer diameters;[7] Au nanorice;[8] Ag nanoprisms synthesized in DMF;[9] plasmonic "dimer" consisting of two adjacent Au nanodisks;[10] silver nanoprisms patterned with colloidal lithography;[11] Ag nanocubes;[12] Au nanostars;[13] and Langmuir–Blodgett Ag nanowire monolayers.[14] Center: far-field scattering image of individual Ag nanoparticles.[15]

widely varying shapes and sizes has been a theme of major importance. As shown in Figure 1,[4–15] a range of chemical methods have been used to generate noble metal nanoparticles of a surprisingly large variety of shapes. The inclusion of surfactants and copolymers along with the chemical reduction of gold or silver acids directs the growth of gold or silver onto preferential crystalline facets of the nanoparticle, resulting in the growth of solid, highly crystalline nanoparticles of various shapes. Other growth methods involve the deposition of noble metals onto a dielectric supporting nanoparticle, covering it partially to form a semishell, for example, or completely to form nanoparticles such as nanorice. Synthesis of silver–gold nanostructures, followed by the sacrificial removal of silver, results in the synthesis of hollow gold nanoparticles whose morphology depends on the geometry of the sacrificial Ag nanostructure. Unusual and highly irregular morphologies such as nanostars or branched structures also result from the presence of surfactants in solution-based growth, where crystal twinning creates even more complex morphologies. In addition, substrate patterning methods have been developed, usually by combining chemical and clean-room techniques in unusual ways. Colloidal lithography, where a hexagonally close-packed array of polystyrene nanoparticles is formed followed by the evaporation of noble metal films onto the structure, results in patterned surfaces with nanometer-scale detail over large areas, not achievable by conventional nanofabrication methods. Finally, the tools of clean-room fabrication such as e-beam lithography have also been used to create noble metal disks and islands to examine the role of interparticle spacing in the plasmonic properties of the nanoparticle complex systematically and quantitatively. Together, these structures and many more[16–41] begin to form the toolbox that has shaped our understanding of the properties of localized plasmons in nanoparticles and other nanometer scale metallic structures.

This proliferation of nanoparticles has directly stimulated the development of methods to probe the optical properties of metallic nanostructures at the single particle level. Methods such as dark-field microscopy and other single-particle microscopies reveal detailed properties of nanostructures not detectable by ensemble methods.[13,42–49] Interest in this topic has been intensified by the development of nanostructures as substrates for localized surface plasmon resonance (LSPR) sensing[27,50–52] and for surface-enhanced spectroscopies,[16,53–59] where highly localized "hot spots" give rise to very large spectroscopic enhancements. Understanding which features of nano-

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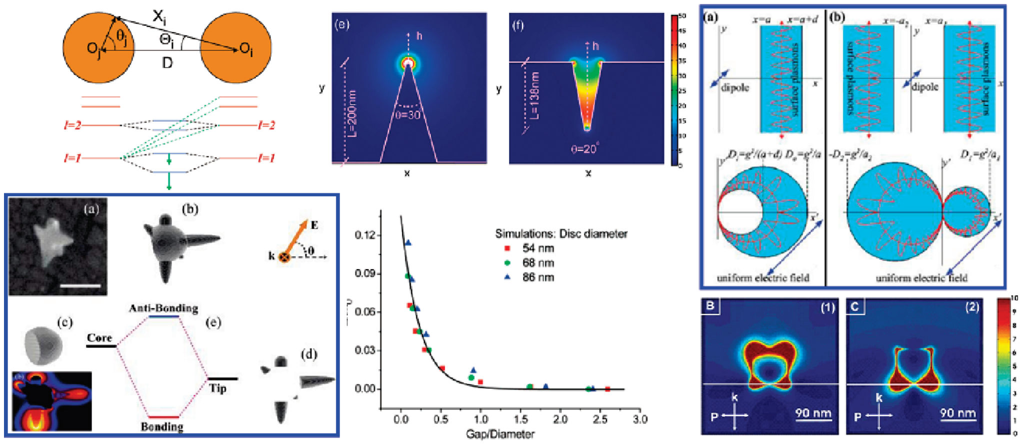

FIGURE 2. Theory for plasmonic systems appearing in *Nano Letters*. Left, top: plasmon hybridization (PH) diagram of the mode energies of a nanoparticle dimer.[60] Left, bottom: PH theory combined with FDTD to identify the plasmonic modes of a nanostar.[61] Center, top: FDTD simulations of the cross-sectional profile of propagating plasmons on nanowaveguides.[62] Center, bottom: universal scaling law for coupling of adjacent plasmonic nanodisks.[10] Right, top: Transformation optics; conformal mapping applied to surface plasmons for ultrabroadband light harvesting.[64] Right, bottom: theoretical studies (FDTD) of localized plasmons of an Ag nanocube[65] reveal the effect of a substrate on its plasmon modes.

structures and their assemblies give rise to large LSPR shifts and strong spectroscopic signals for molecules bound to their surfaces has been a major topic of interest for plasmonics within nanoscience.

Theoretical Insight. The ability to apply classical electromagnetism to describe most of the properties of surface plasmons in metallic structures has been central to the rapid increase in activity in this field. The availability of fast numerical solvers of Maxwell's equations using approaches such as the finite-difference time domain (FDTD), discrete dipole approximation (DDA), finite element (FEM), and boundary element methods (BEM) have enabled the rapid analysis of the properties of complex metallic nanostructures using realistic material parameters and in realistic, anisotropic dielectric environments. While numerical modeling can provide accurate descriptions of plasmonic properties, a unifying theoretical understanding that provides intuitive insight for experimental design is also critically important. Highlights of various theoretical results and approaches are shown in Figure 2.[10,60–65]

The realization that localized plasmon modes mix and hybridize in direct and rigorous analogy with the wave functions of simple quantum systems led to the development of the method of plasmon hybridization (PH). PH provides an intuitive and highly useful picture for understanding the interactions between plasmon modes in coupled nanostructures.[66–68] For example, for two directly adjacent coupled plasmonic nanoparticles the PH method explains both the dramatic color shift and the large electromagnetic enhancement in the junction between two adjacent nanoparticles. Because of this analysis, coupled pairs of nanoparticles are generally referred to as "dimers" with the naming of more complex nanoparticle aggregates, such as trimers, quadrumers, and oligomers following suit.[69] While the PH picture is an analytical method, it can be combined with numerical calculations for the assignment of plasmon modes and the understanding of local fields in highly complex nanostructures, like nanostars.[61] In such complex structures for example, numerical modeling alone would not provide an underlying understanding of the energy dependence of tip plasmon excitation. In contrast, for propagating plasmon systems such as plasmonic nanowaveguides numerical studies have been essential in the understanding of near-field profiles, multimode coupling, and loss.[62] Other theoretical approaches have been used in the analysis and design of plasmonic structures and systems. On the basis of experimental measurements, scaling laws such as a "plasmon ruler equation" have been determined, describing the distance dependence of plasmon coupling in a universal manner.[10] Other approaches from analytical electromagnetism have been adapted to address theoretical problems in plasmonics. Transformation optics, conformal mapping applied to subwavelength optical structures, is an approach that provides a useful method for predicting the properties of nanostructures with unusual optical properties.[64,70–72] Numerical methods, when suitably applied, provide a direct image of the plasmon near field; in systems where reduced symmetry lifts the plasmon mode degeneracy, the resulting distortion of the plasmon modes can be clearly visualized. Fully quantum mechanical approaches[73] have been used to model nanostructures in regimes where classical methods do not faithfully describe their plasmonic properties, for example, in the case of two nearly touching nanoparticles.[74] Theoretical methods provide a range of productive and highly accessible approaches to modeling plasmonic systems of various complexity[34,75–79] with realistic metals

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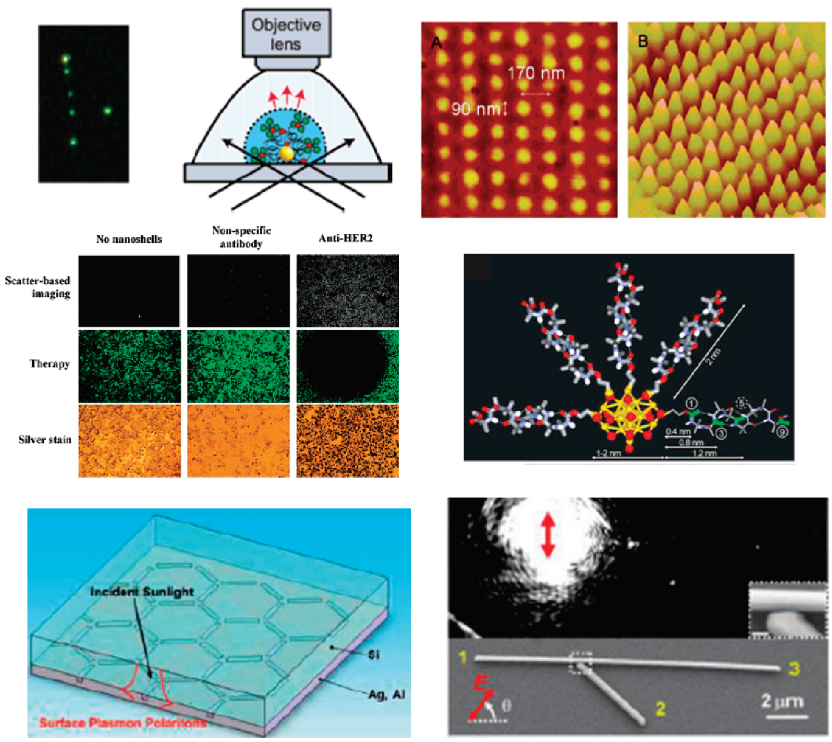

FIGURE 3. A broad range of plasmonics applications appearing in *Nano Letters*. Left top: biosensing with individual plasmonic nanoparticles.[80] Left center: photothermal cancer therapy.[81] Left bottom: plasmonic enhancement of silicon photovoltaic devices using Ag "nanotrenches" that redirect light back into the active device.[82] Right top: plasmonic nanolithography.[83] Right center: a nanoscale molecular thermometer designed to measure plasmon heating.[63] Right bottom: a silver nanowire network serves as a controllable plasmon router.[84]

and in potentially anisotropic environments, which can greatly aid experimental design and interpretation.

Potential Applications. The combination of fabrication methods and powerful theoretical modeling tools has allowed for rapid advances in the field of plasmonics. One result has been a strong and growing interest in applications (Figure 3).[63,80–84] Plasmonics is perhaps the topical area within nanoscience most likely to yield commercial applications in the not-too-distant future that may impact technology and society in significant ways. What is most striking about the potential applications of plasmonics is the breadth of the proposed applications across many disciplines. Just as photonics has penetrated most all modern technologies, we can envision nanoscale optics—plasmonics in a parallel fashion, providing many nanometer-scale practical solutions and contributing in multiple ways to future technologies.

In the first decade of *Nano Letters*, many proof-of-concept applications for plasmonics have been demonstrated, seeding and stimulating further work. One key area, garnering significant interest across the broader scientific community and beyond, has been biotechnology and biomedicine. While gold colloid-antibody conjugates have been used in bioimaging and immunohistochemistry for quite some time, the advent of nanoscience has greatly expanded interest in the development of these systems for applications. A major area of interest has been biosensing.[11,15,26,50,51,80,85–88] The use of gold nanoparticles as robust, unbleachable optical markers for biological assays, essentially replacing bleachable markers such as molecular fluorescent reporters, provides sizable improvements in detection sensitivity. While other bright markers, such as semiconductor nanocrystals, are available, the existence of robust bioconjugation protocols for antibodies to gold nanoparticle surfaces can be directly exploited to develop assays with high affinities. Changes in plasmonic signals due to nanoparticle aggregation provide strong signals for detection, and the ability of plasmonic nanoparticles to selectively enhance or quench fluorescence of nearby molecules[75,89–96] can also be useful in bioassay designs. Monitoring the LSPR shifts of the nanoparticle directly upon binding of an analyte provides a direct and label-free method for bioassays. Since LSPR sensing is essentially a miniaturization of widely used SPR sensing for bioassays, it has attracted a great deal of interest. The challenge of designing nanoparticles with ultrasensitive LSPR sensitivities, to simplify the detection of few-molecule quantities of an analyte, is a highly active area of research.

In addition to in vitro applications such as assays, applications for plasmonic nanoparticles in vivo have garnered extraordinary interest, primarily in the field of cancer research. The large resonant cross section of plasmonic nanoparticles along with their strong photothermal response has been exploited for the light-induced destruction of cancer cells and tumors,[12,81,97–99] a topic

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currently being pursued in clinical trials. The light-scattering properties of plasmonic nanoparticles have also been exploited in this application to visualize the uptake of nanoparticles within tumor tissue using light scattering. As more and more researchers pursue this promising, drug-free therapeutic approach, important variants of this original strategy are being developed.

There are also many exciting applications, as promising as those in the biomedical realm, for contributing significant and potentially transformative advances to current technologies.[25,28,75,83,84,100–104] A prime application of broad scientific and public interest is solar energy harvesting. In photovoltaic cells, the issue of light absorption in the active area of the device is critical for enhancing light-to-electricity conversion efficiencies. This is particularly important for high performance devices, which often have extremely optically thin active areas, to eliminate carrier recombination and to maintain optimum electrical properties. For ultrathin devices, capturing as much light as possible in the active region, thus generating as many carriers as possible, is a critical challenge. One promising strategy is the development of plasmonic groovelike structures fabricated into the back face of the device. The presence of these structures provides a mechanism for backscattering of light transmitted through the device, increasing carrier generation. The nanogrooves scatter light over a large cone of angles back into the device, providing greater efficiencies than would be obtainable by specular back-reflection.

The near field of plasmonic structures focuses light to small volumes and high intensities; this property can be used to expose and develop photoresist, creating plasmon-based nanolithography.[83] The size resolution obtainable by this approach is comparable to the resolution of deep UV or electron-beam lithography, but using plasmonic masks and visible light would provide a far more cost-effective strategy for writing small structures lithographically.

A major characteristic of plasmonic systems is heat generation; because of their large optical cross section and low quantum yield, they are unparalleled as photothermal converters. While numerous applications exploit this property, developing a way to accurately measure the temperature directly at the nanometer scale is an important challenge.[63] Nanoscale thermometry will be important in the advancement of plasmonic applications as well as other areas of research in nanoscience, such as nanocatalysis.

The idea that a propagating plasmon can be used to transmit information has been an important concept since the inception of plasmonics, and many studies have been performed on plasmonic waveguides and active devices. Ag nanowires have emerged as an important nanoscale structure for the subwavelength focusing and transmission of plasmons with only modest loss, such that a variety of plasmonic propagation characteristics can be demonstrated and studied in detail.[105,106] New functionalities such as polarization-dependent plasmon routing can be demonstrated in branched plasmonic nanowires.[84] With studies of photon–plasmon coupling in these geometries, more functional structures are likely to emerge as nanoscale analogs of integrated photonic devices.

Because of the highly diverse nature of this field and its applications, one can be confident that this field will maintain a healthy rate of growth and activity for years to come.

Plasmonics: The Next Ten Years. While inherently risky, it is also quite useful to speculate and make general predictions regarding the future of a field. There are several currently emerging trends that indicate some of the major directions for plasmonics over the next decade. Because of the highly diverse nature of this field and its applications, one can be confident that this field will maintain a healthy rate of growth and activity for years to come. Important new directions for this field include the following: (1) merging plasmonics with quantum systems,[74,79] providing challenges for both theory and experiment; (2) coherent phenomena in plasmonics, where Fano resonances, superradiance, and plasmon-induced transparency result in novel new lineshapes and plasmonic properties;[107,21] (3) active plasmonic devices and media; new types of devices and media combining plasmonics with other functional materials for active and nonlinear responses;[100] (4) improved sensors and detectors; from ultrasmall detectors to LSPR sensors with single molecule sensitivity and specificity,[51] there are many possibilities for advancing the ability to increase detection sensitivities and responsivities; (5) the role of plasmons in modifying chemical reactions; and (6) biomedical applications, where new types of nanoscale devices can be developed for diagnosis[11] and treatment of diseases, to improve treatment efficacy, and to develop prevention strategies for global health challenges. We predict the future of this field to be an exciting one, and we also predict that many of its key advances will be featured on the pages of this journal.

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