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Introduction: Plasmonics and its Building Blocks

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Plasmonics is the area at the intersection of science and technology in which the interaction of light with matter is mediated by surface electromagnetic excitations at a dielectric-metal interface—surface plasmon polaritons and localized surface plasmons. These excitations are collective oscillations of the electrons in the vicinity of a planar vacuum-metal interface, or in a metallic particle with nanoscale dimensions. They can be generated by illuminating a suitably designed dielectric-metal interface by light. The frequency of the resulting excitations can be made to match that of the incident light, but their wavelength can be significantly shorter. The electromagnetic fields of these excitations are localized to well within a wavelength of the interface, with the result that their excitation produces a significant enhancement of the electromagnetic field in the immediate vicinity of the interface. All of these features of surface plasmon polaritons and localized surface plasmons have made them attractive candidates for incorporation into devices for information processing and transmission, and biochemical sensing, that are faster and smaller than existing photonics devices, and faster and at least comparable in size to their electronic counterparts. Thus, they can bridge the size gap between photonics and microelectronic devices.

In this chapter we present an overview of the building blocks of plasmonics, surface plasmon polaritons, and localized surface plasmons, and describe some of their properties that are interesting from a basic science standpoint, and from the potential they show for, or have already shown in, applications. It is intended to serve as an introduction to topics that will be discussed in much greater detail in the remaining chapters of this book.

1.1 Surface Plasmon Polaritons

A surface plasmon polariton is an electromagnetic wave that, in its simplest form, propagates along the planar interface between a dielectric and a metal with an amplitude that decays exponentially with increasing distance into each medium from the interface. If the metal is lossy, the amplitude of this wave also decreases exponentially in the direction of its propagation.

Such surface electromagnetic waves were first discussed by Zenneck in 1907 [1.1] who showed that Maxwell's equations together with the corresponding boundary conditions admit a solution that is a surface wave. The electric field of this wave is p polarized (TM polarized), i.e. its magnetic vector is perpendicular to the sagittal plane, namely the plane defined by the direction of propagation of the wave and the normal to the surface. In the case that the dielectric medium, characterized by a real positive dielectric

constant ϵ_0 occupies the half space $x_3 > 0$, and is in contact across the plane $x_3 = 0$ with a medium characterized by a frequency-dependent dielectric function $\epsilon(\omega)$ that occupies the half space $x_3 < 0$, the nonzero component of the magnetic field of this wave propagating in the x_1 direction can be written as

$$H_2(x_1, x_3|\omega) = \exp[ikx_1 - \beta_0x_3] \quad x_3 > 0, \quad (1.1a)$$

$$= \exp[ikx_1 + \beta x_3] \quad x_3 < 0. \quad (1.1b)$$

A time dependence $\exp(-i\omega t)$ has been assumed in writing this field, but is not indicated explicitly. The functions β_0 and β , the inverse decay lengths of the field into the medium with ϵ_0 and the medium with ϵ , respectively, are given by

$$\beta_0 = \left[k^2 - \epsilon_0(\omega/c)^2 \right]^{\frac{1}{2}}, \quad (1.2a)$$

$$\beta = \left[k^2 - \epsilon(\omega)(\omega/c)^2 \right]^{\frac{1}{2}}, \quad (1.2b)$$

where ω is the frequency of the wave. From the boundary conditions on the field at the interface $x_3 = 0$ the wavenumber k is found to be

$$k(\omega) = \frac{\omega}{c} \left[\frac{\epsilon_0\epsilon(\omega)}{\epsilon_0 + \epsilon(\omega)} \right]^{\frac{1}{2}}. \quad (1.3)$$

When this result is substituted into the expressions (1.2a) and (1.2b) for the inverse decay lengths, the latter become

$$\beta_0(\omega) = \frac{\omega}{c} \left[\frac{-\epsilon_0^2}{\epsilon_0 + \epsilon(\omega)} \right]^{\frac{1}{2}}, \quad (1.4a)$$

$$\beta(\omega) = \frac{\omega}{c} \left[\frac{-\epsilon^2(\omega)}{\epsilon_0 + \epsilon(\omega)} \right]^{\frac{1}{2}}. \quad (1.4b)$$

If the preceding results are to describe a surface wave, we see from Eq. (1.1) that $Re\beta_0$ and $Re\beta$ must both be positive. This is generally achieved in one of two ways. We can assume that $\epsilon(\omega)$ is real and negative. For historical reasons that will be discussed below, the resulting surface electromagnetic wave has become known as a Fano mode. Its phase velocity is smaller than the speed of light in the dielectric medium with the dielectric constant ϵ_0 .

Alternatively, we can assume that $\epsilon(\omega)$ is complex, $\epsilon(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega)$ with an imaginary part $\epsilon_2(\omega)$ that is non-negative. In this case we find from Eq. (1.3) that the wavenumber $k(\omega)$ is now complex, $k(\omega) = k_1(\omega) + ik_2(\omega)$, where both $k_1(\omega)$ and $k_2(\omega)$ are real. The energy propagation length of the resulting surface wave is then given by

$$\ell(\omega) = (2k_2(\omega))^{-1}. \quad (1.5)$$

There are now two cases to consider. In the case of a weakly lossy medium for which

$$\epsilon_2(\omega) \ll |\epsilon_1(\omega)|, \quad \epsilon_2(\omega) < \epsilon_0, \quad (1.6)$$

we find from Eq. (1.3) that

$$k_1(\omega) = \frac{\omega}{c} \left[\frac{\epsilon_0 |\epsilon_1(\omega)|}{|\epsilon_1(\omega)| - \epsilon_0} \right]^{\frac{1}{2}} \times \left\{ 1 - \frac{\epsilon_2^2(\omega)}{8} \frac{\epsilon_0 [4|\epsilon_1(\omega)| - \epsilon_0]}{\epsilon_1^2(\omega) [|\epsilon_1(\omega)| - \epsilon_0]^2} \right\}, \quad (1.7a)$$

$$k_2(\omega) = \frac{\omega}{2c} \epsilon_2(\omega) \frac{\epsilon_0^{3/2}}{|\epsilon_1(\omega)|^{\frac{1}{2}} [|\epsilon_1(\omega)| - \epsilon_0]^{3/2}}, \quad (1.7b)$$

to the first nonzero order in $\epsilon_2(\omega)$. In writing these expressions we have assumed that $\epsilon_1(\omega)$ is negative. The corresponding inverse decay lengths become

$$\beta_0(\omega) = \frac{\omega}{c} \frac{\epsilon_0}{[|\epsilon_1(\omega)| - \epsilon_0]^{\frac{1}{2}}} \left[1 + \frac{i}{2} \frac{\epsilon_2(\omega)}{|\epsilon_1(\omega)| - \epsilon_0} \right], \quad (1.8a)$$

$$\beta(\omega) = \frac{\omega}{c} \frac{|\epsilon_1(\omega)|}{[|\epsilon_1(\omega)| - \epsilon_0]^{\frac{1}{2}}} \left\{ 1 + i \frac{\epsilon_2(\omega)}{2} \frac{2\epsilon_0 - |\epsilon_1(\omega)|}{|\epsilon_1(\omega)| [|\epsilon_1(\omega)| - \epsilon_0]} \right\}, \quad (1.8b)$$

also to the first nonzero order in $\epsilon_2(\omega)$. From these results we see that if $\epsilon_1(\omega) < -\epsilon_0$, the real parts of $\beta_0(\omega)$ and $\beta(\omega)$ are positive, so that the corresponding electromagnetic wave is localized to the interface. Because $\beta_0(\omega)$ and $\beta(\omega)$ are complex quantities it is called a ‘‘generalized’’ surface wave. It can be regarded as a Fano mode, weakly attenuated by ohmic losses in the metal.

Let us now consider a very lossy metal for which $\epsilon_2(\omega)$ satisfies the inequalities

$$\epsilon_2(\omega) \gg |\epsilon_1(\omega)|, \quad \epsilon_2(\omega) \gg \epsilon_0. \quad (1.9)$$

In this case we find from Eq. (1.3) that the real and imaginary parts of the wavenumber $k(\omega)$ are given by

$$k_1(\omega) = \sqrt{\epsilon_0} \frac{\omega}{c} \left\{ 1 - \frac{\epsilon_0}{2\epsilon_2^2(\omega)} \left[\epsilon_1(\omega) + \frac{3}{4}\epsilon_0 \right] \right\}, \quad (1.10a)$$

$$k_2(\omega) = \sqrt{\epsilon_0} \frac{\omega}{c} \frac{\epsilon_0}{2\epsilon_2(\omega)}. \quad (1.10b)$$

The corresponding inverse decay lengths are

$$\beta_0(\omega) = \epsilon_0 \frac{\omega}{c} \frac{1}{[2\epsilon_2(\omega)]^{\frac{1}{2}}} (1 + i), \quad (1.11a)$$

$$\beta(\omega) = \frac{\omega}{c} \left[\frac{\epsilon_2(\omega)}{2} \right]^{\frac{1}{2}} (1 - i), \quad (1.11b)$$

whose real parts are positive. The electromagnetic wave described by these results is therefore bound to the surface, but because $\beta_0(\omega)$ and $\beta(\omega)$ are complex quantities it is also a generalized surface wave.

We see from Eq. (1.10a) that if $\epsilon_1(\omega) < -(3/4)\epsilon_0$ at the frequency of the surface wave, $k_1(\omega)$ lies to the right of the light line $k = \sqrt{\epsilon_0}(\omega/c)$, and the generalized surface wave described by Eqs. (1.10) and (1.11) is just a damped surface wave that is attenuated as it propagates with an energy mean free path $\ell(\omega) = (c/\omega)\epsilon_2(\omega)/\epsilon_0^{3/2}$. However, if $\epsilon_1(\omega) > -(3/4)\epsilon_0$, $k^{(1)}(\omega)$ lies to the left of the light line, and we have a new type of electromagnetic surface wave, namely the surface wave studied by Zenneck [1.1]. In particular, Zenneck modes can exist even when $\epsilon_1(\omega)$ is positive, provided the inequalities (1.9) are satisfied. The imaginary part of $\epsilon(\omega)$ gives rise even in this case to inverse decay lengths $\beta_0(\omega)$ and $\beta(\omega)$, Eqs. (1.11), whose real parts are positive, i.e. to an electromagnetic wave bound to the interface $x_3 = 0$. The phase velocity of these waves is therefore greater than the speed of light in the medium whose dielectric constant is ϵ_0 . It is readily seen from Eq. (1.4) that a surface wave cannot exist if $\epsilon(\omega)$ is real and positive.

From the earliest days following their discovery Zenneck waves were controversial. Two years after Zenneck's work Sommerfeld [1.2] studied the electromagnetic field excited by an oscillating vertical dipole above a planar conducting surface, and concluded that this field goes over into that of a cylindrical Zenneck wave near the surface at large distances from the dipole. However, there was a sign error in Sommerfeld's analysis that he himself discovered and corrected [1.3], and which was rediscovered subsequently by Norton [1.4, 1.5], that invalidated this result. In addition, in several other theoretical studies of the electromagnetic field excited in this manner [1.6–1.8] no result showing the excitation of a Zenneck wave was obtained. The conclusions drawn from these studies are that in an intermediate range surrounding the dipole and close to the surface, the field excited is essentially that of a cylindrical Zenneck wave, but with increasing radial distance r along the surface from the source it becomes proportional to $1/\sqrt{r}$, a dependence characteristic of wave propagation in two dimensions [1.9].

The experimental observation of Zenneck waves was hindered by the fact that their phase velocity is greater than that of light, which prevented the use of the method of attenuated total reflection or of a grating for this purpose. An additional difficulty encountered in efforts to detect these waves is the fact that any real source at the interface produces an electromagnetic field that consists of both bulk and surface waves. Separating these two contributions is a difficult experimental task [1.10, 1.11]. Nevertheless, Zenneck waves excited by other methods have now been observed experimentally in several different situations: on saltwater in the microwave frequency range [1.12], on a dielectric surface in the optical frequency range [1.13], and on a metal surface in the terahertz frequency range [1.14]. In spite of this experimental evidence for the existence of Zenneck surface waves, some theoretical questions appear to remain concerning these waves, in particular concerning their excitation by sources at the interface. Histories of the studies of Zenneck waves are presented in Refs. [1.15, 1.16].

In the context of plasmonics Zenneck surface electromagnetic waves appear to have a significant potential for applications based on surface waves in the gigahertz and terahertz frequency ranges, when the metallic surface supporting them is periodically structured.

The earliest manifestation of a surface plasmon polariton in experimental data, although it was not recognized as such until much later, occurred in 1902. In measurements of the angular and wavelength dependencies of the reflectivities of light diffracted from various metallic gratings, Wood noted “anomalies” in the data he obtained [1.17, 1.18]. These anomalies were of two types, and occurred when the magnetic vector of the incident light was parallel to the grooves of the grating, i.e. in p polarization. The first type of anomaly was a discontinuous change of intensity along the spectrum at well-defined wavelengths that were independent of the metal from which the grating was formed, and were determined by

the period of the grating. These anomalies were explained by Lord Rayleigh [1.19, 1.20] as occurring at the wavelengths at which a diffracted order appears or disappears at a grazing angle. For the n th diffracted order the corresponding wavelength is given by $\lambda_n = a(1 - \sin \theta_0)/n$, where a is the period of the grating, θ_0 is the angle of incidence, and n is an integer. In the former case the power in that order is removed from the zero-order beam; in the latter case the power in that order is returned to the zero-order beam. The result in either case is a critical angle change in the specular reflectivity.

It should be noted that such anomalies also occur at the Rayleigh wavelengths when the electric vector of the incident light is parallel to the grooves of the grating, i.e. in s polarization, but they are weak and require deep grooves for their observation [1.21, 1.22].

The second type of anomaly, now called a Wood's anomaly, was diffuse, and extended in a wide interval of wavelengths from a Rayleigh anomaly toward longer wavelengths, and generally consisted of a maximum and minimum of intensity. These anomalies were found to occur only in p polarization, when the plane of incidence was perpendicular to the grooves of the grating, and their spectral positions changed when the dielectric function of the metal changed. Wood had no explanation for these anomalies. The explanation for them was provided in a seminal paper by Fano [1.23], who showed that they are due to the excitation of the surface plasmon polaritons supported by a periodically corrugated vacuum-metal interface by the incident light. Since the wave number of a surface plasmon polariton, Eq. (1.3), is slightly larger than ω/c , i.e. larger than the component of the wave vector of the incident light parallel to the surface, the difference between them is made up by the wavenumber of the grating, $2\pi n/a$, which enables the conservation of momentum in the interaction of the incident wave with the surface plasmon polariton. Thus the condition for the excitation of the surface plasmon polariton is $k = k_{sp}(\omega) + (2\pi/a)n$, or equivalently, $(1/\lambda) = (1/\lambda_{sp}(\omega)) + n/a$. Since surface plasmon polaritons exist only in p polarization, they cannot be excited by s-polarized light when its electric vector is parallel to the grooves of the grating. Hence Wood's anomalies do not exist in s polarization. Since the wavenumber (wavelength) of a surface plasmon polariton is a function of the dielectric function of the metal, the scattering angles or wavelengths at which these anomalies occur vary from metal to metal.

In the years that followed Fano's work surface electromagnetic waves were studied primarily by engineers seeking to use periodically corrugated metallic surfaces or planar metal surfaces coated by a dielectric film, as waveguides, particularly in the microwave region of the electromagnetic spectrum [1.24–1.27].

Interest in surface electromagnetic waves began to revive in the late 1950s. Measurements of the characteristic electron energy loss spectra of aluminum and magnesium by Powell and Swan [1.28–1.30] showed that the loss spectrum of each metal displayed two peaks. The higher frequency peak was identified with the loss due to the excitation of the bulk plasmon of each metal, predicted by Pines and Bohm [1.31], while the lower frequency peak was identified with the loss due to the excitation of the surface plasmon supported by each metal, predicted by Ritchie [1.32], and in subsequent work by Stern and Ferrell [1.33].

Theoretical interest in surface electromagnetic waves began to grow in the 1970s [1.34] and 1980s [1.35, 1.36]. However, this work was concerned primarily with basic physical properties of these surface waves of various types. Possible applications of these waves were little discussed, if at all.

A stimulus to the study of surface electromagnetic excitations was provided by the experimental discovery of surface enhanced Raman scattering [1.37, 1.38] and the enhancement of second harmonic generation in reflection from a metal surface [1.39]. It was soon recognized that surface roughness, by

enhancing the strength of the electromagnetic field at points on the metal surface, was responsible for much of the observed enhancements. These observations stimulated investigations into the enhancement of the electromagnetic fields associated with small metallic particles of various shapes, which will be discussed in Section 1.2, as well as with localized topographical defects [1.40], with periodically corrugated surfaces [1.41, 1.42], and with randomly rough surfaces [1.43].

Although the confinement of the electromagnetic field to the near vicinity of planar-dielectric-metal interfaces, and its enhancement thereby, had been known for some time, its use in devices for information processing and information transmission took some time to be realized. This appears to be due to the existence of competing technologies for such applications, namely electronics, and then photonics, which was replacing electronics in various applications. The need for ever faster information processing led to the fabrication of smaller and smaller electronic and photonic integrated components, so that more of them could be integrated on a chip, for example. It was when the size of these components began to approach the length scale of electron and light waves that a search for a new approach to the nano-miniaturization of device components began. In the case of optical components ways had to be found to overcome the diffraction limit, which states that it is not possible to focus or confine a three-dimensional light beam to a lateral size that is smaller than half of its wavelength in the host medium. For light in the visible to near infrared region of the optical spectrum, i.e. with wavelengths in the 400–1000 nm range, the diffraction limit forbids the creation of dielectric optical elements with lateral dimensions smaller than approximately 200 nm. In the case of electronic components, as their size becomes smaller than the mean free path of the electrons in a metal, which at room temperature is of the order of a few tens of nanometers [1.44], the scattering of electrons from the boundaries of these components has to be taken into account in their design, as well as the fact that the electron energy levels become discrete rather than essentially continuously distributed. The localization of the electromagnetic field of a surface plasmon polariton to within strongly subwavelength distances from a dielectric metal interface provides a means for overcoming the diffraction limit, and creates the ability to consider the fabrication of devices of nanoscale dimensions.

The resulting studies of properties of surface plasmon polaritons that might make them useful for incorporation into nanoscale devices, and the fabrication of structures for experimental investigations of these properties, have been greatly aided by the availability of computers with the great speed and large memories required for solving the electromagnetic boundary value problems that arise in the design of devices. The use of computational methods such as the finite-difference time-domain approach, the finite element method, the discrete dipole approximation, and others, in obtaining solutions to these problems was a significant step in enabling numerical solutions to be obtained. At the same time, new fabrication techniques such as ion beam milling, electron beam lithography, and self-assembly, have been developed that enable micro- and nano-structures to be created. All of these developments have produced an explosion of interest in this field of research, now known as plasmonics.

1.2 Localized Surface Plasmon Resonances

Although many of the theoretical and experimental studies in the area of plasmonics, as well as the applications stemming from these studies, have been carried out for surface plasmon polaritons on metallic surfaces that are planar in the absence of any structuring, these are not the only surface excitations of interest in this field.

At about the time that Zenneck was carrying out his investigation of surface electromagnetic waves, in a study of the scattering of light by a dielectric sphere Mie [1.45] showed that such a sphere supports electromagnetic resonances at discrete frequencies which, when excited by an incident electromagnetic field, produce significant enhancements of the field in the vicinity of the sphere. The frequencies of these resonances depend on the dielectric function of the sphere and of the medium in which it is embedded, and on its size. Many years later Fröhlich [1.46] studied these resonances in the electrostatic (unretarded) approximation, which is applicable when the diameter of the sphere is much smaller than the wavelength corresponding to the frequency of the resonance being studied. From the solutions of Laplace's equation inside and outside the sphere, and the associated boundary conditions, Fröhlich found that the frequency of the ℓ th resonance ($\ell = 1, 2, 3, \dots$) is obtained from the solution of the equation

$$\frac{\epsilon(\omega)}{\epsilon(\omega) - \epsilon_M} = \frac{\ell + 1}{2\ell + 1}, \quad (1.12)$$

where $\epsilon(\omega)$ is the dielectric function of the sphere, and ϵ_M that of the surrounding medium. Fröhlich's interest was in spheres fabricated from ionic crystals. In the case of a diatomic cubic polar crystal for which the dielectric function is given by $\epsilon(\omega) = \epsilon_\infty(\omega_L^2 - \omega^2)/(\omega_T^2 - \omega^2)$, where ϵ_∞ is the optical frequency dielectric constant of the particle, while $\omega_{L,T}$ are the frequencies of the long wavelength longitudinal and transverse optical phonons, the frequency of the infrared active mode ($\ell = 1$) is $\omega = \omega_T [(\epsilon_0 + 2\epsilon_M)/(\epsilon_\infty + 2\epsilon_M)]^{1/2}$, where $\epsilon_0 = \epsilon_\infty(\omega_L^2/\omega_T^2)$ is the static dielectric constant of the particle.

Equation (1.12) holds for spheres fabricated from materials other than ionic crystals, in particular from metals. Thus, if for $\epsilon(\omega)$ we assume the simple free electron form $\epsilon(\omega) = 1 - (\omega_p^2/\omega^2)$, where ω_p is the plasma frequency of the electron gas, the resonance frequencies obtained from Eq. (1.12) are

$$\omega_\ell = \omega_p \left[\frac{\ell}{(\epsilon_M + 1)\ell + \epsilon_M} \right]^{1/2}. \quad (1.13)$$

The frequency of the dipole-active mode ($\ell = 1$) is therefore given by $\omega_p[1/(2\epsilon_M + 1)]^{1/2}$. At this resonance frequency a strong extinction of light occurs.

We note that the resonance frequencies obtained from Eq. (1.12) are independent of the size of the sphere.

In subsequent work, stimulated by experiments in which optical properties of small free-standing ionic crystal particles of cubic shape were studied [1.47, 1.48], calculations of the frequencies of the resonance modes of cubic particles were carried out [1.49–1.51], as well as the resonance frequencies of particles in the form of rectangular parallelepipeds [1.52]. Calculations of these frequencies have now been carried out for free-standing particles in the form of a prolate spheroid [1.53–1.55], an oblate spheroid [1.53, 1.56], a nanorod [1.57], and two types of nanoshells, namely a metallic sphere coated with a dielectric, and a dielectric sphere coated with a metal [1.58]. Such nanoshells are of particular interest because the frequencies of the localized surface plasmon resonances they support can be tuned for given core and shell materials by varying the radius of the core and/or the thickness of the shell.

For a particular arbitrary shape of a nanoparticle, the equation from which its resonance frequencies can be determined can be obtained from the result that [1.59]

$$\lambda(\omega)\phi^>(\mathbf{x}) = \frac{1}{4\pi} \int_{S_0} dS' G(\mathbf{x}|\mathbf{x}') \frac{\partial}{\partial n'} \phi^>(\mathbf{x}'), \quad (1.14)$$

where $\lambda(\omega) = \frac{\epsilon(\omega)}{\epsilon_M - \epsilon(\omega)}$, $\phi^>(\mathbf{x})$ is the solution of Laplace's equation in the region outside the particle that vanishes at infinity, S_0 denotes the surface of the particle, and $\partial/\partial n$ is the derivative along the normal to the surface S_0 at each point of it directed away from the volume of the particle. The function $G(\mathbf{x}|\mathbf{x}') = |\mathbf{x} - \mathbf{x}'|^{-1}$ is the free-space Green's function that satisfies $\nabla^2 G(\mathbf{x}|\mathbf{x}') = -4\pi\delta(\mathbf{x} - \mathbf{x}')$. When the point \mathbf{x} is allowed to approach the surface S_0 from the region outside the particle, an integral equation eigenvalue problem is obtained. If the electrostatic potential $\phi^>(\mathbf{x})$ outside the particle is expanded according to

$$\phi^>(\mathbf{x}) = \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} \frac{a_{\ell m}}{|\mathbf{x}|^{\ell+1}} Y_{\ell m}(\theta, \phi), \quad (1.15)$$

where $Y_{\ell m}(\theta, \phi)$ is a spherical harmonic, substitution of this expansion into Eq. (1.14) yields the equation satisfied by the expansion coefficient $\{a_{\ell m}\}$ in the form [1.52],

$$\lambda(\omega)a_{\ell m} = \frac{1}{2\ell + 1} \sum_{\ell'=0}^{\infty} \sum_{m'=-\ell'}^{\ell'} N(\ell, m; \ell', m') a_{\ell' m'}. \quad (1.16)$$

The matrix elements $N(\ell, m; \ell', m')$ are defined in terms of an integral over the surface of the particle

$$N(\ell, m; \ell', m') = \int_{S_0} dS \left[|\mathbf{x}|^{\ell} Y_{\ell m}^*(\theta, \phi) \frac{\partial}{\partial n} \left(\frac{Y_{\ell' m'}(\theta, \phi)}{|\mathbf{x}|^{\ell'+1}} \right) \right]. \quad (1.17)$$

In general, these matrix elements have to be calculated numerically. The vanishing of the determinant of the matrix of coefficients in Eq. (1.16) yields the eigenvalues $\{\lambda(\omega)\}$, and hence the resonance frequencies of the particle.

The surface of the particle is necessary for the existence of these resonant modes, which are sometimes called surface plasmon resonances or localized surface plasmons, in the case of metallic particles. However, the frequencies of these modes while depending on the shape of the particle, in the unretarded case are independent of their size [1.59].

1.3 Constructions

Over the years theoretical and experimental research in plasmonics has proceeded in two directions. One is devoted to investigations of properties of surface plasmons and polaritons for basic science reasons, and in the expectation that the more that is known about these surface excitations the wider is the possible scope of their applications. The other is devoted to the study of specific devices based on surface plasmons and polaritons. These directions, however, are not always sharply distinct, and can sometimes merge. As a result of these investigations new types of surface plasmon polaritons and localized surface plasmons have been predicted and realized in the laboratory. At the same time a variety of applications of these building blocks have been proposed, some of which have been tested experimentally. In this section we describe briefly the constructs, both conceptual and tangible, realized through the two lines of investigation. Many of them will be discussed at much greater length in the remaining chapters of this book.

1.3.1 Excitation and Observation of Surface Plasmon Polaritons

In many theoretical studies of properties of surface plasmon polaritons the manner in which the surface wave is created is not described. It is simply present. For experimental studies of surface plasmon polaritons, and for theories of these experiments, methods of exciting surface plasmon polaritons are crucial.

A volume electromagnetic wave incident from vacuum on the planar surface of a metal cannot excite a surface plasmon polariton. Although it is possible for the frequency of the incident field to match the frequency of the surface plasmon polariton, it is not possible to match the tangential component of the wave vector of the incident field to the wave number of the surface plasmon polariton because the latter is in the non-radiative region of the frequency-wavenumber plane.

We have seen earlier in this chapter that p-polarized light whose plane of incidence is perpendicular to the grooves of a metallic grating can couple into the surface plasmon polaritons supported by the grating and excite them to produce the Wood's anomalies in the reflectivity. The first experiments in which surface plasmon polaritons were excited in this manner for the purpose of studying properties of these surface waves, namely the magnitude, direction, and phase of their fields, were carried out by Beaglehole [1.60].

Surface plasmon polaritons excited by light incident normally on a metallic grating whose profile is symmetric propagate equally in opposite directions. If unidirectional propagation of the excited surface plasmon under this condition is desired, an asymmetric grating profile can be used [1.61].

It is not necessary to use an essentially infinitely long grating to excite surface plasmon polaritons. A truncated grating, consisting of a few (four or five) grooves ruled on the metal surface on which the surface plasmon polariton is to propagate, illuminated by a p-polarized volume electromagnetic wave can also be used for this purpose [1.62].

In principle a finite segment of one realization of a one-dimensional randomly rough surface, or even an isolated groove or ridge, could replace the grating and still enable the excitation of a surface plasmon polariton when illuminated by a volume electromagnetic field. However, the excitation efficiencies of these two approaches are lower than that of a grating coupler.

Prism couplers are used to excite surface plasmon polaritons through the phenomenon of attenuated total reflection. In one version of this method [1.63] an electromagnetic field is incident through a prism on the metal surface on which the surface plasmon polariton is to be excited. The base of the prism is parallel to the metal surface, and is separated from it by a gap filled with a medium whose index of refraction is smaller than that of the prism. It is often air. When the angle of incidence in the prism is greater than the critical angle for total internal reflection at the prism-gap interface, at some value of it the evanescent electromagnetic field in the gap region can couple to a surface plasmon polariton, exciting the surface wave thereby. A dip in the angular dependence of the reflectivity of this structure at that angle is the signature of the excitation. A prism coupler of this type can also convert a surface plasmon polariton into a volume electromagnetic wave propagating into the prism away from its interface with the gap region. If such a prism is placed downstream from the prism used to excite a surface plasmon polariton, and the intensity of the field propagating in it is measured as a function of the distance between the two prisms, the energy propagation length of the surface plasmon polariton can be determined [1.64]. This method for exciting a surface plasmon polariton is now known as the Otto attenuated total reflection method.

In a variant of this method [1.65] a thin metal film deposited on the base of a prism is illuminated through the prism at an angle of incidence greater than the critical angle for total internal reflection.

The light reflected from the prism-metal interface induces an evanescent field that can couple to the evanescent fields of the surface plasmon polaritons supported by this structure. In this reflection structure a surface plasmon polariton is bound to both the prism-metal interface and the metal-vacuum interface. However, only the surface wave bound to the metal-air interface is excited by this approach. This method is now called the Kretschmann-Raether method.

Although Otto [1.63] and Kretschmann and Raether [1.65] are generally credited with the introduction of the prism-coupling approach to the excitation and detection of surface plasmon polaritons, it is worth noting that nearly a decade earlier Turbadar [1.66], had used the Kretschmann-Raether geometry in an experimental study of the reflectivity of a thin aluminum film deposited on the base of a glass prism through which the light was incident. His data show a well-defined, very deep, minimum in the reflectivity at an angle of incidence just beyond the critical angle for total internal reflection at the prism-metal film interface. In subsequent work he carried out similar measurements on thin copper, gold, and silver films [1.67], and observed deep minima in the angular dependencies of the reflectivities of these metals at angles of incidence beyond the critical angle as well [1.67]. Unfortunately, Turbadar did not state that this feature was the result of the excitation of a surface plasmon polariton at the metal-air interface. As a result, the credit for prism-coupling excitation of surface plasmon polaritons has been given to Otto and to Kretschmann and Raether.

It should be noted that the periodic corrugations of the metal surface, the presence of a dielectric gap in the Otto method, and the use of a thin metal film in the Kretschmann-Raether method, produce experimental surface plasmon polariton dispersion curves that depart to a greater or lesser extent from the dispersion curve for these surface waves at a planar dielectric metal interface given by Eq. (1.3).

The illumination of a narrow gap between the edge of a razor blade and the metal surface by a volume electromagnetic wave has been used to excite surface plasmon polaritons [1.68].

End-fire coupling [1.69] has been used in an experiment to excite a surface plasmon polariton guided by a thin gold film embedded in SiO_2 [1.70]. It has also been used in theoretical studies of other types of guided surface plasmon polaritons [1.71–1.74]. In this method a surface plasmon polariton at a dielectric-metal interface is excited by a p-polarized volume electromagnetic wave incident on the end face of the dielectric-metal system that is normal to the interface.

Finally, we note that the excitation of surface plasmon polaritons on a gold film in the Kretschmann-Raether attenuated total reflection configuration by optical four-wave mixing has been demonstrated [1.75–1.77]. The metal film is illuminated through a prism by two collinear p-polarized laser beams of frequencies ω_1 and ω_2 . Through the metal's third order susceptibility $\chi^{(3)}$ a nonlinear polarization at the frequency $2\omega_1 - \omega_2$ is produced that excites a surface plasmon polariton at that frequency. The excitation of the surface wave is observed by a dip in the dependence of the four-wave mixing intensity on the angle of incidence. This method of excitation of surface plasmon polaritons differs from earlier methods of nonlinear excitation which used second harmonic generation [1.78–1.80], i.e. through the metal's second order susceptibility $\chi^{(2)}$, for this purpose.

Methods for observing surface plasmon polaritons are fewer in number than methods for exciting these surface electromagnetic waves. The earliest technique used for measuring the local surface plasmon polariton field on a surface with nanometer resolution was scanning tunneling microscopy (STM) [1.81], in which the additional tunneling current induced by surface plasmon polaritons was detected.

However, the proximity of a metal tip to the metal surface supporting the surface plasmon polariton significantly perturbs the surface plasmon polariton field being measured [1.82]. This makes such tips ill-suited for the direct measurement of the surface plasmon polariton field on a metal surface.

Scanning near-field optical microscopy (SNOM), in which uncoated optical fiber tips kept at a fixed distance of a few tens of nanometers above a metal surface are scanned across it, provides a way of measuring the surface plasmon polariton field above the surface with a minimal perturbation of that field [1.83]. The signal detected with such a probe is closely proportional to the intensity of the electric field at its position [1.84]. In this mode of operation both the field and the surface topography are measured simultaneously, which is useful in studying surface plasmon polaritons on structured or otherwise perturbed surfaces.

Although optical fiber tips coated with a metal layer have been used in SNOM measurements, primarily of surface topographies, [1.85], in application to the determination of surface plasmon polariton fields they suffer from the same drawback as do the metal tips used in STM, namely they significantly perturb the field being measured.

The opening angle of the optical fiber probe tip plays an important role when it is the field of a surface plasmon polariton scattered by a surface defect that is sought. In such a scattering process some of the energy in the incident surface wave is converted into volume electromagnetic waves in the vacuum above the surface. The discrimination of the field of the latter waves from the evanescent field of the incident and scattered surface plasmon polaritons is facilitated by the use of a probe tip with a wide opening angle [1.86].

1.3.2 Guiding of Surface Plasmon Polaritons

In order to use surface plasmon polaritons in applications it is necessary to be able to guide them. Ideally, one would like to find guiding mechanisms that restrict the lateral confinement of the surface wave to subwavelength dimensions, and guide it not only along straight lines but also around corners, or in configurations such as Y-splitters and S-bends. In practice the lateral confinement is the result of a compromise, because the smaller the lateral confinement the shorter is the propagation length of the surface wave. Nevertheless, several mechanisms for guiding surface plasmon polaritons have been proposed, and several of them have been implemented in the laboratory. These include metal films of finite width (metal stripes), which can be deposited on a dielectric, e.g. glass, substrate [1.87], or embedded in an infinite homogeneous dielectric medium [1.88]; a triangular metallic wedge on a planar surface of the same metal [1.89]; a dielectric-loaded surface plasmon polariton waveguide that consists of a dielectric ridge, usually of a rectangular cross section placed on a metal surface [1.90, 1.91]; a channel cut into an otherwise planar metal surface [1.92, 1.93]; a planar metal surface on which is deposited a doubly periodic array of nano particles in which a channel is created by the removal of one or more adjacent parallel rows of nano particles [1.94]; and a trench cut into a metal film on a dielectric substrate and coated with a polymer [1.95] or air [1.96], or into a free-standing metal film [1.97].

Several of these guides produce surface plasmon polariton propagation distances of several tens of microns, with subwavelength lateral confinement.

1.3.3 Attenuation of Surface Plasmon Polaritons and its Suppression

As a surface plasmon polariton propagates along a dielectric-metal interface it is attenuated. This attenuation decreases its energy propagation length, and can degrade the performance of devices based on these surface electromagnetic waves. As a result ways of overcoming this attenuation or increasing the mean free path have been sought.

A major mechanism for the attenuation of a surface plasmon polariton as it propagates along even a planar dielectric-metal interface is ohmic losses in the metal. Methods for increasing the propagation distances of a surface plasmon polariton include incorporating a metallic thin film into a multilayer structure in which it is surrounded symmetrically [1.98,1.99] or asymmetrically [1.100,1.101] by dielectric layers. In this way one of the two surface waves supported by the film can be made to have a weak electric field inside the film, where the ohmic losses occur, increasing its propagation length thereby. The resulting electromagnetic surface waves have come to be known as *long-range surface plasmon polaritons*.

Another mechanism for overcoming the effects of absorption on surface plasmon polariton propagation is to place the metal surface supporting the surface wave in contact with a gain medium [1.102,1.103]. The presence of the gain medium can compensate the absorption losses in the metal, leading to an increased propagation length of the surface plasmon polariton. An experimental demonstration of the complete suppression of loss by this mechanism has been carried out for a surface plasmon polariton propagating at the interface between a silver film and an optically pumped polymer with a dye [1.104]. When the gain exceeds the loss, amplification of the surface wave occurs. Indeed, the first experimental measure of gain in propagating surface plasmon polaritons was carried out on the long range surface plasmon polariton in a structure consisting of a gold stripe on a SiO₂ substrate covered by a gain layer consisting of optically pumped dye molecules in solution [1.105]. The ability to amplify surface plasmon polaritons has the potential for applications in several fields, e.g. for improving the performance of biosensors [1.106].

A second mechanism for the attenuation of surface plasmon polaritons is surface roughness. As a surface plasmon propagates on a randomly rough surface it is attenuated by two mechanisms: (i) roughness-induced conversion into volume electromagnetic waves in the vacuum region, propagating away from the surface and (ii) roughness-induced scattering into other surface plasmon polaritons. Both of these mechanisms produce attenuation by scattering out of the incident beam. The dispersion of the surface wave is also affected by the surface roughness which in general produces wave slowing.

The dispersion and damping of surface plasmon polaritons on one-dimensional randomly rough metal surfaces has been calculated in Ref. [1.107]. In this work it was shown that the random roughness leads to wave slowing, binds the surface wave closer to the surface than a planar surface does, and produces an energy propagation length that is shorter on a lossless randomly rough metal surface than it is on a lossy planar metal surface.

The dispersion [1.108,1.109] and damping [1.108–1.111] of surface plasmon polaritons on two-dimensional randomly rough surfaces have been studied by several authors, with results that are qualitatively similar to those obtained for one-dimensional randomly rough surfaces.

Experimental measurements of the attenuation of surface plasmon polaritons on randomly rough surfaces that can separate the contribution to it from the ohmic losses and roughness-induced losses have yet to be carried out.

1.3.4 Surface Plasmon Polariton Scattering from Surface Defects

A surface plasmon polariton incident on a surface defect is partly scattered into other surface plasmon polaritons and is partly converted into volume electromagnetic waves in the vacuum above the surface. The ability to control the propagation of surface plasmon polaritons is central to their use in devices. It is therefore important to be able to calculate the cross sections for these two types of scattering processes.

The scattering out of the beam by both processes decreases the propagation length of a surface plasmon polariton, and it is important to be able to estimate the magnitude of this decrease. At the same time surface defects of particular forms and sizes can scatter surface plasmon polaritons in specified desirable ways, e.g. they can act as mirrors for surface plasmon polaritons [1.112] or as flashlights [1.113, 1.114], and can focus them as well [1.115, 1.116].

The scattering of surface plasmon polaritons by surface defects of various types has been studied theoretically by several approaches. The majority of these calculations have been carried out for scattering from one-dimensional surface defects, namely grooves and ridges, or for parallel arrays of such defects. Thus, scattering from an isolated groove or ridge [1.117–1.120] or from a finite periodic array of grooves or ridges [1.121] was studied on the basis of a simple impedance boundary condition [1.122]. A more sophisticated impedance boundary condition was used in subsequent studies of scattering from an isolated groove or ridge, or from an impedance defect [1.123, 1.124]. The reduced Rayleigh equation was also used in calculations from an isolated groove or ridge [1.125]. A modal expansion method has been used to study the scattering of a surface plasmon polariton by a finite periodic array of grooves ruled on a metal surface [1.126, 1.127]. Two different Green's function approaches have been applied to the solution of this problem, namely a surface integral method [1.128] and a volume integral method [1.129–1.131]. Finally, finite-difference time-domain and boundary-element methods have been used to calculate the scattering of a surface plasmon polariton from a deep groove [1.132]. The results of these calculations show, for example, how the shape and size of a groove or ridge can be chosen to produce a volume electromagnetic field in the vacuum region that propagates away from the surface at a specified angle of scattering, or to maximize the reflectance of a surface plasmon polariton at the expense of the transmittance and/or the conversion into volume waves.

The scattering of a surface plasmon polariton by a localized two-dimensional surface defect has been less intensively studied. The scattering of a surface plasmon polariton by a circularly symmetric indentation (dimple) on an otherwise planar metal surface, and from a circularly symmetric protuberance formed from the same metal as the substrate, has been calculated by a rigorous, purely numerical, nonperturbative solution of the corresponding reduced Rayleigh equation [1.133]. The results for scattering into other surface plasmon polaritons show a shadow region behind the defect, and a well-defined maximum in the angular dependence of the intensity of the volume electromagnetic field in the vacuum which occurs for in-plane scattering at a moderate value of the polar scattering angle, namely $\theta_s = 28^\circ$. The scattering from a dielectric defect in the form of a rectangular parallelepiped on the planar surface of a metallic film in the Kretschmann-Raether total attenuated reflection geometry [1.65] has been studied by a rigorous Green's tensor approach [1.134]. The scattering from a dielectric defect whose shape is an anisotropic Gaussian or an anisotropic hemiellipsoid on the planar surface of a metal has been calculated with the use of an effective boundary condition [1.135]. Finally, the scattering from a circularly symmetric dielectric defect of Gaussian form on a planar metal surface has been calculated by a rigorous, purely numerical, nonperturbative solution of the corresponding reduced Rayleigh equation [1.136]. The results show that the scattering into other surface plasmon polaritons is primarily in the forward direction. There is no shadow region behind the defect. The maximum in the angular dependence of the volume electromagnetic field scattered into the vacuum occurs in in-plane scattering at a significantly larger polar scattering angle $\theta_s = 70^\circ$, than the angle at which it occurs in the scattering from a circular symmetric dimple in a planar metal surface [1.133]. Its position is independent of the aspect ratio of the Gaussian defect and of its dielectric constant. Thus, there are qualitative and

quantitative differences between the scattering of surface plasmon polaritons from localized metallic and dielectric defects.

1.3.5 Accelerating Surface Plasmon Polaritons

A new class of surface plasmon polaritons has been discovered recently, namely *accelerating surface plasmon polaritons*. Their discovery was inspired by earlier work by Berry and Balazs [1.137], who showed that the one-dimensional time-dependent Schrödinger equation for a particle propagating in free space has a non-spreading solution expressed in terms of the Airy function [1.138]. It moves with a velocity that increases linearly with time, and hence is an accelerating beam. This solution, however, has an infinite energy content.

It was soon recognized that the equation solved by Berry and Balazs is equivalent to the equation for the propagation of a two-dimensional scalar optical field in the paraxial approximation. This field is nondiffractive, and its intensity maximum propagates along a parabolic trajectory. Thus, the acceleration of this beam is not related to time, as it is for the Berry-Balazs solution, but describes the deviation of the Airy beam from a straight, forward, propagation direction. This beam also contains infinite energy.

A finite energy Airy beam was derived by Siviloglou and Christodoulides [1.139] within the paraxial approximation, and an experimental realization of this beam was carried out by Siviloglou *et al.* [1.140] soon after. Shortly after those discoveries a new type of accelerating beam was predicted on the basis of the paraxial approximation, namely the parabolic beam [1.141], whose experimental observation was reported soon after [1.142].

Because the Airy and parabolic beams were obtained on the basis of the paraxial approximation to a scalar wave equation, the transverse acceleration of these beams is always restricted to small angles. Consequently attention shifted toward overcoming the paraxial limit of accelerating beams. Several have now been found. These include a circular nonparaxial Airy beam [1.143–1.145], a beam that propagates along a circular arc [1.146–1.148], a Mathieu beam [1.146], and a Weber beam [1.146]. The latter beams bend through large angles along elliptic and parabolic trajectories, respectively. The circular [1.149], and the Mathieu, and Weber beams [1.133], have been realized in the laboratory.

It was inevitable that surface plasmon polariton analogs of nondiffracting accelerating beams would be sought. Salandrino and Christodoulides [1.150] predicted Airy surface plasmon polaritons in a paraxial approximation. These plasmonic beams are almost diffractionless, possess an indifference to surface perturbations, and propagate along parabolic trajectories. Their manipulation by linear optical potentials was studied theoretically not long after [1.151]. Several experimental methods for generating plasmonic Airy beams, and for studying their properties, have demonstrated their self-bending property, their nearly diffraction-free propagation, and their robustness against surface perturbations, such as surface roughness [1.152–1.154].

Accelerating plasmonic beams may well find applications in optical interconnects and manipulation of nanoparticles on metal surfaces. Their ability to propagate around corners without the need for structuring the surface with plasmonic waveguides is an effect that should be exploitable.

1.3.6 Surface Plasmon Polariton Lasers

To create a laser one needs a gain medium and a feedback mechanism that forces light to travel forward and backward through the gain medium until a threshold value of its intensity is reached [1.155].

In conventional lasers the feedback is provided by partially transparent mirrors. However, in the early 1970s it was shown that laser oscillations can be obtained by the use of the guided modes of a planar film waveguide, in which a light-amplifying dye was dissolved in the film to provide the gain, and where the feedback was provided by the backward intramode diffraction from a spatially periodic modulation of the film's complex refractive index [1.156]. It was also found that laser oscillations can be obtained on the basis of the guided modes of a leaky planar film waveguide where a dye provides the gain and the backward intramode diffraction from a spatially periodic modulation of the film's thickness provides the feedback [1.157].

Many years later Capasso and his co-workers [1.158] created laser waveguides emitting in the far infrared that were based on surface plasmon polaritons at a two-metal grating deposited on top of a semiconductor quantum cascade active material [1.159]. The latter provides the gain, while the former produces the feedback.

Li *et al.* [1.160] have demonstrated that a structure consisting of a two-dimensional periodic array of nanoscale holes piercing an aluminum oxide layer deposited on an aluminum substrate displays the attributes of a surface plasmon polariton laser in the visible region of the optical spectrum when the holes are filled with a gain medium (fluorescein). The array of holes allowed light to be coupled to and from the structure and provided feedback to the surface plasmon polaritons.

In subsequent work by this group [1.161], the aluminum oxide layer deposited on an aluminum substrate and pierced by a doubly periodic array of nanoscale holes was covered by graphene. Gain was provided by core/shell CdSe/ZnS quantum dots that were deposited on the graphene layer. The feedback to the surface plasmon polariton was provided by the same periodic array of holes that enabled the incident light to couple to the surface plasmon polaritons. This structure displayed the attributes associated with an optically pumped surface plasmon polariton laser in the visible region of the optical spectrum.

1.3.7 Transformation Optics

A powerful tool for guiding the flow of light in a medium in nearly arbitrary ways is transformation optics [1.162, 1.163]. This approach exploits the form invariance of Maxwell's equations under a transformation of coordinates to produce electric permittivity and magnetic permeability tensors of a medium that force light in the medium to propagate along specified trajectories. The resulting medium in general is anisotropic and inhomogeneous. This creates difficulties for the fabrication of such materials. Nevertheless, this method has been used to design a variety of optical devices, including invisibility cloaks [1.162, 1.163], beam bends [1.164], field rotators [1.165, 1.166], field concentrators [1.165, 1.166], beam shifters [1.167, 1.168], and beam splitters [1.168]. Most of these devices have not been fabricated yet.

In recent work the method of transformation optics has been applied to plasmonics to control the flow of surface plasmon polaritons on dielectric-metal interfaces [1.169–1.174]. The application of the transformation optics method to plasmonics problems at a dielectric-metal interface is not without difficulties, because the expressions for the electromagnetic material parameters should in principle be implemented both in the dielectric and in the metal. This poses significant technical challenges: because the penetration depth of a surface plasmon polariton into the metal is subwavelength, the electric permittivity and magnetic permeability tensors within the metal would need to be manipulated on a nanometer scale. Nevertheless, transformations of the material properties in both media have been used by Kadic *et al.* [1.171] to design an invisibility cloak, a field concentrator, and a rotator for surface

plasmon polaritons. However, it has been shown that under certain conditions the transformation optics approach can be simplified by carrying out the coordinate transformation only in the dielectric medium, leaving the metal unchanged [1.169, 1.170, 1.172, 1.173]. In addition it has been shown that if only linear coordinate transformations are used, the resulting transformed electromagnetic material tensors become anisotropic but homogeneous, which simplifies the fabrication of the corresponding metamaterials [1.171, 1.174]. An interesting plasmon-related application of transformation optics is to the highly accurate determination of the van der Waals interaction between two metallic microspheres and between a metallic microsphere and a plane [1.175]. This interaction, which can be obtained from the zero-point energy of the electrostatic modes of these structures, plays a significant role in a variety of physical phenomena such as friction [1.176], surface adhesion [1.177], and nanoparticle self-assembly [1.178]. Accurate calculations of this interaction are difficult, so a method that yields such results is very welcome.

Surface plasmon polariton devices that have been designed by the transformation optics method include invisibility cloaks [1.169, 1.171, 1.173, 1.174, 1.179, 1.180], a field concentrator [1.171, 1.174], a guider [1.174], a rotator [1.174], a beam shifter [1.169, 1.173], a bend [1.170, 1.173], a lens [1.173], and Luneberg and Eaton lenses [1.170, 1.172]. Of these only the surface plasmon polariton cloak [1.180], Luneberg lens [1.172], and Eaton lens [1.172] have been fabricated.

An alternative approach to the cloaking of a surface structure from observation by a surface plasmon polariton that is not based on transformation optics is provided by a structure that consists of two concentric rings of equally spaced scatterers [1.181]. The polarizabilities of the scatterers are determined iteratively in a least squares sense so as to annul the total electric field within the circular array, while leaving the field outside it unchanged. A surface structure inside the circular array becomes invisible to an incident surface plasmon polariton.

1.3.8 Plasmonic Metamaterials

Not long after a medium (metamaterial) characterized by a dielectric permittivity and a magnetic permeability that are both negative within some frequency range was first realized experimentally [1.182], the propagation of surface electromagnetic waves on such a medium began to be studied [1.183–1.187]. It was found that these waves have interesting properties. For example, one and the same surface can support surface plasmon polaritons of both p- and s-polarization in non-overlapping frequency regions, and the energy flux and the phase velocity of these waves are oppositely directed.

These double-negative, or negative index, media are not the only metamaterials on which the propagation of surface electromagnetic waves has been studied. It has also been studied on hyperbolic media [1.188, 1.189]. These are anisotropic materials for which, for example, the dielectric constant ϵ_{zz} is positive, while ϵ_{xx} and ϵ_{yy} are negative, or *vice versa*. Their name derives from the fact that the surfaces of constant frequency for waves in these media are hyperbolic, instead of spherical, as for an isotropic medium. Such a material can be fabricated relatively easily [1.189]. One approach to their fabrication is to create multilayer structures from alternating layers of a metal and a dielectric whose thicknesses are much smaller than the operating wavelength. The surface waves studied in Ref. [1.188] occur at the interface between a metal and a hyperbolic medium in the case that the optical axis of the latter lies in the interface. The electromagnetic field in the metal is a superposition of evanescent p- and s-polarized waves, not just a p-polarized wave. Depending on the frequency range these surface waves can propagate in all directions on the interface, or only into a narrow range of angles.

Closely related to hyperbolic media are indefinite media [1.190]. These are anisotropic media in which not all of the principal components of the electric permittivity and the magnetic permeability tensors have the same sign. The propagation of surface electromagnetic waves along the planar interface between an indefinite medium and a conventional medium, which can be a metal, has been studied theoretically in Ref. [1.190]. Conditions for the existence of such waves have been obtained, and it has been shown that surface waves of both p and s polarization can exist at such an interface.

The interface between a metal and an anisotropic metamaterial the component of whose electric permittivity normal to the interface is close to vanishing, an epsilon-near-zero metamaterial, can support above-light-line surface plasmon polaritons [1.191]. In turn these modes induce fast-wave non-radiative modes which leads to a wide-angle perfect absorption in an ultra thin epsilon-near-zero metal bilayer.

A material that has attracted significant interest in the context of plasmonics is doped graphene [1.192]. This is a monolayer of carbon atoms arranged in a honeycomb lattice. A single graphene sheet can support a p-polarized surface plasmon propagating on it [1.193–1.195]. In the limit of large wave numbers the frequency of this wave is proportional to the square root of its wavenumber, as in the case of a wave on a two-dimensional electron gas [1.196]. The same graphene sheet can also support an s-polarized surface plasmon propagating on it in a narrow frequency range [1.193, 1.197]. However, this wave is only weakly bound to the graphene sheet. More strongly bound s-polarized surface plasmons are supported by a graphene bilayer, namely two parallel graphene sheets separated by a dielectric layer [1.198, 1.199], or if the graphene sheet is deposited on a nonlinear dielectric substrate [1.200]. The propagation of p-polarized surface plasmons on a graphene bilayer has also been studied [1.201]. The frequencies of graphene surface plasmons typically fall in the terahertz frequency range, but their dispersion and propagation characteristics can be varied by changing the gate voltage or the strength of a magnetic field oriented normally to the graphene sheet [1.195, 1.201]. An extensive review of theoretical studies of surface plasmons on graphene monolayers and bilayers is contained in the article by Bludov *et al.* [1.202]. Experimental studies of graphene surface plasmons by near-field microscopy [1.203, 1.204] have produced images of their fields, determinations of their wavelengths, and have demonstrated altering the amplitude and wavelength of these surface waves by changing the gate voltage.

Other types of metamaterials of interest in plasmonics exist. An important class of such materials consists of structured surfaces. These include metallic surfaces formed from periodically arranged spherical voids created by electrochemical deposition through a template of self-assembled latex spheres [1.205], surface plasmon polaritonic crystals [1.206], namely doubly periodic arrays of scatterers on a planar metal surface, and randomly rough surfaces [1.207]. Many of the properties and applications of these and other structured surfaces are described in a recent book [1.208].

A related type of metamaterial that is proving to be useful in plasmonics is a periodically corrugated perfectly conducting surface. It mimics a metal surface in the gigahertz and terahertz frequency ranges, and supports surface electromagnetic waves that resemble surface plasmon polaritons [1.209, 1.210]. A planar metallic surface does not support surface plasmon polaritons in these frequency ranges, only surface skimming bulk waves. Such surfaces can guide these low frequency waves, and are finding applications in biochemical sensing [1.211], pharmaceutical quality control [1.212], and in security screening [1.213].

Transparent conducting oxides make possible high-performance metamaterial devices operating in the far infrared [1.214]. Transition metal nitrides can substitute for metals at visible frequencies [1.215].

The creation of metamaterials with, in many cases, tailored electromagnetic properties, gives rise to new types of surface electromagnetic surface waves that have the potential for use in applications.

1.3.9 Nanoparticles on Substrates

Resonance frequencies and field distributions of nanoparticles of various shapes just above or deposited on a planar substrate have been calculated. The modes of a metal sphere above a substrate have been studied as functions of its distance above the substrate [1.215–1.218]. This geometry arises in theoretical studies [1.202, 1.203] of light emission from tunnel junctions formed by depositing gold particles on an anodized aluminum film [1.219].

The modes of nanoparticles on a substrate have been studied for particles of several different shapes: a sphere [1.218], a truncated sphere [1.220], a hemisphere [1.40], a hemispheroid [1.54], a cube [1.221], a nanoshell [1.222], a truncated tetrahedron [1.223], an ellipsoid [1.224], and a nanorod [1.225]. Although some of these calculations were carried out in the context of some physical problem, e.g. infrared absorption by ionic crystals [1.40] and surface enhanced Raman scattering [1.54], the majority of them were performed to determine how the presence of the substrate affects the resonance frequencies of the variously shaped nanoparticles obtained in its absence.

These frequencies are shifted from the values they have in the absence of the substrate, and degeneracies of modes are partially or completely lifted by the lowered symmetry of the system in which the nanoparticle now finds itself. For example, the triply degenerate $l = 1$ mode of a spherical nanoparticle is split into two modes, one nondegenerate and the other doubly degenerate, when the particle is close to a planar substrate.

In addition, the interaction of a plasmonic nanoparticle with a nearby metallic substrate leads to the hybridization of the localized surface plasmon of the nanoparticle and the propagating surface plasmon polaritons of the adjacent metal surface. It has been shown [1.225] that this hybridization is the electromagnetic analog of the hybridization of a localized electronic state with a continuous band of electronic states, which is described by the spinless Fano-Anderson model [1.226]. An interesting consequence of this hybridization occurs when the nanoparticles form a doubly periodic lattice on the metallic substrate. The dispersionless localized surface plasmon resonance frequency can coincide with a frequency on the dispersive sinusoidal-like dispersion curve of the surface plasmon polaritons of the substrate. The dispersion curve of the system then displays a repulsion of levels in the vicinity of the point where the two curves cross in the absence of the interaction. This effect has been observed experimentally [1.227].

When the substrate is a dielectric instead of a metal, this interaction of the plasmonic nanoparticle with it is weak: the nanoparticle couples only to its image in the dielectric, screened by the factor $(\epsilon - 1)/(\epsilon + 1)$, where ϵ is the dielectric constant of the substrate [1.228].

1.3.10 Surface Shape Resonances

Somewhat related to localized surface plasmon resonances associated with a metallic particle close to or on a substrate are electrostatic and electromagnetic surface shape resonances. These are obtained from solutions of Laplace's and Maxwell's equations, respectively, that are spatially localized in the vicinity of a topographical surface defect, such as a protuberance or indentation on an otherwise planar metal surface in contact with a dielectric, often vacuum. The frequencies of these modes, and the field distributions associated with them, depend on the shape of the protuberance or indentation.

In the electrostatic limit the resonance frequency of ridges and grooves defined by a Lorentzian surface profile function has been calculated [1.229, 1.230], as well as of ridges and grooves on a metal film deposited on a planar dielectric substrate [1.231]. The resonance frequencies corresponding to an electrostatic potential that is even in the coordinate normal to the generators of the surface are lower than $\omega_p/\sqrt{2}$, the frequency of a surface plasmon on a planar metal surface, in the case of a ridge, and are higher in the case of a groove. The reverse is the case for the modes corresponding to an electrostatic potential that is odd in this coordinate.

The frequencies of the resonance modes of two-dimensional, circularly symmetric, protuberances and indentations with shapes that include a hemisphere [1.232], and a hemisphere on a planar surface [1.233], have also been calculated. These shapes allow the use of coordinate systems in which Laplace's equation separates, and in which the surface of the protuberance or indentation is a constant coordinate surface. Other circularly symmetric surface profile functions for which the frequencies of the surface shape resonances they support have been calculated include a Gaussian [1.230], a Lorentzian [1.230], an exponential [1.230], and a Gaussian profile on a metal film on a planar dielectric substrate [1.234]. In these cases the frequencies corresponding to a protuberance lie below $\omega_p/\sqrt{2}$, while those corresponding to an indentation lie above $\omega_p/\sqrt{2}$.

1.3.11 Fabrication of Nanoparticles

Several methods have been used to fabricate nano particles of various shapes. Most of these nanoparticles have been fabricated from gold and silver and their alloys, because their localized surface plasmon resonances occur in the visible region of the electromagnetic spectrum.

Silver nanocubes on a glass substrate [1.221, 1.235] have been produced by means of the polyol synthetic method [1.236], in which silver nitrate is reduced with ethylene glycol in the presence of poly (vinyl pyrrolidone) (PVP). Gold nanoboxes have also been fabricated by this method [1.235]. These are hollow gold polyhedra bounded by six {100} facets and eight {111} facets.

Arrays of triangular nanoparticles have been made by the use of nanosphere lithography [1.237]. In this method an ordered array of polystyrene nanospheres is formed on a substrate. A metal is then deposited onto the substrate through the interstices between the nano spheres. The spheres are then removed, leaving behind an array of metallic particles.

Single gold triangular prisms were produced [1.238] on a glass substrate by seed-mediated growth [1.239]. In this method gold seed particles are first prepared in a mixture of NaBH_4 and H_2O -DMF containing PVP and HAuCl_4 . The seeds are then added to a growth solution consisting of aqueous HAuCl_4 and PVP, and complete reduction of the gold is then allowed to occur. The resulting solution after purification contains mainly trigonal prisms and smaller numbers of nanoparticles of other shapes.

Citrate-capped gold nanoparticles obtained by reduction of HAuCl_4 with borohydride ions serve as seeds for the growth of gold nanorods by the seed-mediated growth method [1.240]. Improved versions of this approach to the fabrication of gold nanorods have been reported in Refs. [1.241, 1.242]. Nanorods with aspect ratios of up to 10 have been fabricated by the last cited approach.

Nanoshells with Au_2S cores and gold shells have been created by mixing aqueous solutions of HAuCl_4 and Na_2S [1.243]. Gold nanospheres have also been produced in this way [1.243]. Nanoshells with gold cores and silica shells [1.244] have been fabricated by a modified version of a method used to produce luminescent core-shell silica nanoparticles [1.245].

A variant of a nanoshell consisting of a spherical dielectric core coated with a metallic layer is nanorice [1.246]. This nanoparticle, which consists of a prolate spheroid dielectric core, coated by a metallic layer has been fabricated by the approach used in metallizing the dielectric cores of nano shells [1.243].

Nanoeggs, in which the center of a spherical dielectric core is shifted from the center of the metallic shell surrounding it, have been fabricated by anisotropic deposition of additional metal onto previously fabricated dielectric metal concentric nanoshells [1.247].

Gold nanorings have been produced on soda-glass substrates by colloidal lithography [1.248]. In this method a thick gold film is evaporated on a self-assembled array of polystyrene colloidal particles deposited on the glass substrate. Ion beam etching is used to remove the gold film, during which secondary sputtering of materials creates a gold shell around the sides of the polystyrene particles. The polystyrene particles are removed by a UV-ozone treatment and a water rinse, producing free-standing gold nanorings. A slight variant of this process has been used to produce gold nanodisks [1.248].

1.3.12 Some Applications of Nanoparticles

A major application of metallic nanoparticles and nanoshells is in optical biosensing [1.249–1.251]. The resonance frequency of a localized surface plasmon is extremely sensitive to the dielectric constant of the medium surrounding it. Changes in the dielectric constant of this medium, even by the attachment of a single molecule to the metallic nanoparticle produces a shift of its surface plasmon resonance frequency that is measurable by present day optical techniques [1.250]. The localized surface plasmon response is not chemically specific. However, approaches to overcoming this disadvantage have been developed [1.252, 1.253].

The electromagnetic field scattered from a rough metal surface or from a metallic nanoparticle can be significantly enhanced relative to the incident electromagnetic field. Some of this enhancement has chemical origins but the largest part of it is electrodynamic in origin. Two processes contribute to the latter effect. The first is the “lightning rod” effect, namely the enhancement of the electromagnetic field at a sharp metallic tip. The second is the excitation of localized surface plasmons at the metal surface. This electromagnetic field enhancement produces an enhancement of the otherwise weak Raman signal from molecules adsorbed on a rough metal surface by up to a factor of 10^6 in intensity [1.37, 1.38]. The spectroscopic method that has arisen from this effect, surface enhanced Raman spectroscopy (SERS), is now widely used because it provides information about molecular vibrations, for example, which may be useful for chemical identification, that other spectroscopic methods cannot provide. A great deal of effort is now being put into developing and optimizing SERS substrates to achieve large enhancements. Enhancement factors of about 10^{10} have been obtained for molecules on films formed from gold and silver nanoshells deposited on a glass substrate [1.254]. In this case it is the excitation of the localized surface plasmon resonances supported by the nanoshells that is responsible for this enhancement.

The extremely large cross sections achievable in surface enhanced Raman scattering have made it possible to study single molecules on single nanoparticles at room temperature, in the visible [1.255] and near-infrared [1.256] frequency regions. The ability to carry out such studies with high sensitivity is of great scientific and practical interest in chemistry, biology, medicine, pharmacology, and environmental science [1.257].

To increase solar harvesting in organic solar cells plasmonic silver nanoparticles of uniform size were deposited on transparent electrodes [1.258]. The increased photocurrent density caused by the

enhanced absorption of the photoactive conjugate polymer by the large electromagnetic field strength in the vicinity of the excited localized surface plasmons increased the overall power conversion by 23%.

Linear chains of metallic nanoparticles have been used as plasmonic waveguides [1.259].

A significant application of metallic nanoparticles is to the fabrication of optical nanoantennas. These can take several forms. They can consist of one-dimensional [1.260] and two-dimensional [1.261] arrays of specially shaped individual particles, arrays of pairs of particles [1.262–1.264], and arrays of nanoshells [1.265], that convert propagating electromagnetic fields into localized or confined electric fields, and *vice versa*. These structures can be two-dimensional in nature, such as metal stripes [1.266], or three-dimensional, such as nanorods [1.263], bow tie particles [1.267], nano discs [1.268], and spheres [1.269]. In this way the large mismatch between optical wavelengths in the visible (hundreds of nanometers) and much smaller nanoscale objects can be largely overcome.

The strongly subwavelength field confinement and the strong field enhancement in the near field of the nanoparticles, depend on the size and shape of the particles, and in the case of particle pairs, on their separation. These features have led to important applications of optical nanoantennas [1.270]. These include biological sensing, where the presence of a small molecule near to a nano antenna can produce a detectable change in its spectral response [1.271], single molecule sensing [1.272], optical communications [1.273], high-resolution microscopy [1.274], and single-photon and single-plasmon sources [1.275].

The study and utilization of optical nanoantennas is a rapidly developing area of plasmonics, with a great potential for additional applications in basic science and technology.

Nanoparticles offer the possibility of being used in the treatment of cancer. For example [1.276], when the resonance frequency of a nanoshell is tuned to the near infrared, a frequency region where optical absorption in tissue is minimal and penetration is optimal [1.277], and they are injected into a tumor, their strong absorption of moderately low exposures of externally applied near-infrared light delivers a dose of heat that kills the cancer cells without harming the surrounding healthy tissue.

Additional applications of plasmonic nanoparticles are described in other sections of this chapter.

1.3.13 Quantum Plasmonics

The majority of the theoretical studies in plasmonics to date have been carried out on the basis of classical electrodynamics or electrostatics, namely on the basis of Maxwell's or Laplace's equations and the corresponding boundary conditions. However, in recent work attention has begun to be directed toward aspects of plasmonics involving the interaction of light with matter in which the quantum nature of either the light or the matter has to be taken into account, and leads to effects not obtainable by classical electromagnetic theory. This interest has given rise to the subfield of quantum plasmonics.

An example of where the quantum nature of the incident light has to be taken into account is provided by the excitation of a single quantum of a surface plasmon polariton on a metallic nanowire by a single photon from an isolated quantum dot [1.278]. An example of where the quantum nature of the matter with which the incident light interacts is found in the dependence of the frequency of a localized surface plasmon resonance with decreasing particle size. A typical electron mean free path at room temperature is 30 nm [1.44]. This implies that in the design of electronic devices with smaller lateral dimensions the fact the mean free path of an electron is larger than the size of a metallic nanoparticle has to be taken into account. In this case the scattering of an electron from the boundary of the nanoparticle

is important in determining its dielectric function. Moreover, the energy levels of an electron in the particle are now quantized and discrete. These effects lead to a shift in the localized surface plasmon resonance frequency from the value predicted on the basis of the Drude model for the dielectric function of the particle. Recent experiments [1.279] carried out on individual silver nano spheres showed that as their diameters decreased from 20 to 2 nm the resonant frequency shifted to higher energy by 0.5 eV, in agreement with the results of a quantum-mechanical calculation of the particle's dielectric permittivity.

Yet another example of where the quantum nature of a plasmonic effect is manifest is provided by a spaser (surface plasmon amplification by stimulated emission). The concept of a spaser was introduced in 2003 by Bergman and Stockman [1.280] (see also Refs. [1.281, 1.282]). In its simplest form a spaser is a metallic nanoparticle surrounded by, or in close proximity to, a gain medium. The nanoparticle, the spaser's core, supports localized surface plasmon resonances. The size of the nanoparticle can be just a few nanometers, much smaller than any wavelength in the system, so that the plasmonic eigenmodes supported by it can be treated in the quasistatic approximation, rather than by means of electrodynamics. The electric field strength associated with the excitation of one of these plasmonic modes can be very large, $\sim 10^6$ V/cm. The gain medium overlaps the spasing surface plasmon field spatially, and its emission line overlaps this mode spectrally. In the case that the gain medium is a dye, a pump beam excites electron-hole pairs in the gain medium, which relax to form excitons. The excitons recombine, emitting surface plasmons in the nanoparticle. The plasmons create the high local electric fields that excite the gain medium and stimulate more emission to this mode, which is the feedback mechanism. If the feedback is strong enough, and the lifetime of the spaser mode is long enough, stimulated emission of the surface plasmons occurs. Outcoupling of the surface plasmon oscillations to photonics modes then produces lasing.

A spaser has some similarity to a conventional laser, but it does not emit photons. Instead it emits localized surface plasmons, with the nanoparticle supporting these modes playing the role of the laser's resonant cavity. As in the case of a laser the energy source for the spasing mechanism is a gain medium that is excited externally, for example by a laser pulse.

In view of these similarities it is not surprising that proposals for the creation of a nano laser on the basis of the spaser concept were advanced by several groups [1.283, 1.284], and in fact several groups have now constructed spaser-based nano lasers.

Noginov *et al.* [1.285], used a system of nanoparticles formed from a gold core (14 nm diameter), which provided the localized plasmon modes, surrounded by a silica shell (15 nm thickness) containing an organic dye, which provided the gain, in an aqueous suspension. The surface plasmon resonance band for this nanoparticle corresponds to a wavelength $\lambda \sim 520$ nm, which overlaps both the excitation and emission bands of the dye molecules. This system was pumped at a wavelength $\lambda = 485$ nm with ~ 5 ns pulses. A peak in the stimulated emission spectrum of this system is observed at $\lambda = 533$ nm. A series of tests showed that the observed phenomenon, which combined resonant energy transfer from excited dye molecules to surface plasmon oscillations, and stimulated emission of surface plasmons in the form of light, is consistent with the original theoretical proposal of a spaser [1.280].

A desire to overcome the ohmic losses that are present in the metallic components of nanoscale devices, and generally require their operation at low temperatures, has led to attention being directed at spasers that operate at room temperature. Thus, Ma *et al.* [1.286] have created a semiconductor plasmon laser that operates at room temperature. It consists of a 45 nm thick, 1 μ m length cadmium sulfide (CdS) nanosquare separated from a silver substrate by a 5 nm thick magnesium fluoride (MgF_2) layer. The

proximity of the high permittivity CdS square to the silver substrate allows the transverse magnetic modes of the former to hybridize with surface plasmon polaritons at the MgF₂-silver interface, leading to strong confinement of light in the MgF₂ gap region, with relatively low metal loss. The total internal reflection of surface plasmon polaritons at the boundaries of the gap provides the strong feedback needed for lasing. Experimental results for the integrated light-pump response show the transition from spontaneous emission through amplified spontaneous emission, to full laser oscillation.

A different room-temperature semiconductor spacer operating near the telecom wavelength $\lambda \sim 1.5 \mu\text{m}$ has been fabricated by Flynn *et al.* [1.287]. It consists of a thin (7.5 nm) gold film of finite width embedded in InP. Eight InGaAs quantum wells are distributed symmetrically on each side of the gold film, within the long-range surface plasmon polariton mode profile, which was calculated to extend ~ 150 nm on each side of the gold film. This structure was pumped from above. It produced free-space radiation at a wavelength $\lambda = 12.46 \mu\text{m}$ emitted from the cavity output-facet when the surface plasmon polaritons reach the end of the gold waveguide. The output of the spaser as a function of the peak pump intensity showed that the output increases quasi-linearly above a clear threshold near $60 \text{ kW}/\text{cm}^2$.

Undoubtedly more realizations of a spaser laser will be forthcoming in the future, perhaps one that is pumped by electrical currents instead of by lasers, which could function as a transistor for all-optical information processing circuits [1.288].

Quantum plasmonics is a new field, but one with great promise for advances in both basic science and in applications.

1.4 Conclusions

Plasmonics is undeniably changing optoelectronics. By providing means for concentrating the energy in optical beams in metallic structures of deeply subwavelength dimensions, it has shrunk the size gap between photonics and electronics. This has opened the door to the possibility of integrating enormous numbers of fast nanoscale devices on a single chip, for applications in information processing and transmission. By providing means for producing strong optical field enhancements plasmonics enables the enhancement of such surface phenomena as surface enhanced Raman scattering, second harmonic generation in reflection from a metal surface, nonlinear effects, and applications in biosensing and medicine, for example.

New types of plasmonic materials have been predicted, and methods for realizing them have been devised. New types of surface electromagnetic waves have been discovered and created in the laboratory. New theoretical approaches have been developed and implemented that predict spatially varying optical properties of a dielectric-metal structure that guide surface electromagnetic waves along specified complex paths that arise in surface plasmon polaritonic circuits and in cloaking applications.

The goal of all nanoplasmonic circuits and devices is still far off, but continuing developments in concepts, algorithms, and fabrication, characterization, and detection techniques are bringing it ever closer.

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