Highly confined optical modes in nanoscale metal-dielectric multilayers

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We show that a stack of metal-dielectric nanolayers, in addition to the long- and short-range plasmon polaritons, guides also an entire family of modes strongly confined within the multilayer—the bulk plasmon polariton modes. We propose a classification scheme that reflects specific properties of these modes. We report experimental verification of the bulk modes by measuring modal indices in a structure made of three pairs of silica (∼29 nm)/gold (∼25 nm) layers.

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Nanoscale confinement of light is of great interest for applications in sensing, imaging, all-optical signal processing, and computing. Subwavelength confinement attributed to gap plasmon polaritons (GPPs) has been demonstrated in a thin dielectric layer surrounded by metallic claddings.1 Here we present another solution to subdiffraction confinement of light and show that a stack of metal-dielectric nanolayers guides a family of modes strongly confined within the multilayer—the bulk plasmon polariton (BPP) modes. The bulk modes have very short penetration depth into the claddings even if the claddings are made of dielectric materials. Their modal indices (ratio of the light velocity in vacuum to the phase velocity of a guided mode) are typically large in absolute value and may be both positive and negative.2 We propose a classification scheme that reflects specific properties of BPPs. We verify BPPs experimentally by measuring their modal indices in a structure made of three pairs of silica-gold nanolayers.

When considering light confinement in a waveguide, the modal index \( n' \) (rather than the group index) is of interest because it defines the modal profile. The field penetration length into the cladding with permittivity \( \varepsilon_d \) is \( L_{cl} = \lambda/2\pi \sqrt{n'^2 - \varepsilon_d} \), where \( \lambda \) is the wavelength. For a given \( \varepsilon_d \), the larger modal index leads to shorter penetration length and stronger confinement. This justifies the interest in the high-index modes. In all-dielectric waveguides, the modal index is smaller than the core index, which limits the confinement scale to \( \sim \lambda/7 \) in a silicon-on-insulator waveguide with silicon core \( (n = 3.5) \) and silica or vacuum claddings.3

The surface plasmon polariton4 (SPP) propagating along the interface between media with permittivities \( \varepsilon_m \) and \( \varepsilon_d \) of different sign (metal and dielectric) is an example of a strongly confined mode. Its modal index \( n_{SPP} = \sqrt{\varepsilon_d \varepsilon_m/\varepsilon_m + \varepsilon_d} \) is slightly above the index of the dielectric. In the visible and near-infrared spectral regions, the permittivity of metal is typically negative and \( |\varepsilon_m| \gg \varepsilon_d \), leading to subwavelength field penetration into metal, while the penetration into the dielectric can be as large as several wavelengths. A remarkable exception is the case of resonant SPPs5 when permittivities of materials at different sides of the interface are exactly opposite: \( \varepsilon_m + \varepsilon_d \rightarrow 0 \) and thus \( n' \rightarrow \infty \). In homogeneous media, the field distribution of the resonant SPP is expected to be confined infinitely close to the interface. The actual scale of the field distribution is defined by the applicability of the concept of dielectric permittivity—that is, by the discrete atomic structure of materials. At optical frequencies, the resonant SPPs are possible if the dielectric has a huge optical gain.6

A metallic film of thickness \( d_m \) between dielectric claddings supports SPPs at each metal-dielectric interface. In sufficiently thin films, coupling between individual SPPs leads to the formation of symmetric and antisymmetric film modes known as long- and short-range plasmon polaritons (LRPs and SRPs).7,8 The coupling leads to a splitting of the dispersion characteristics of these modes. The modal index of the LRP, \( n_{LRP} = \sqrt{\varepsilon_d + (\pi m/\lambda)^2 ((\varepsilon_d - \varepsilon_m)\varepsilon_d/\varepsilon_m)^2} \), is reduced compared to that of the SPP approaching the cladding index \( n_d = \sqrt{\varepsilon_d} \). Its penetration into the claddings increases accordingly. The modal index of the SRP is higher than that of the SPP: \( n_{SRP} = \sqrt{\varepsilon_d + (\pi m/\lambda)^2 (\varepsilon_d/\varepsilon_m)^2} \), yielding the confinement scale \( L_{SRP} = d_m + 2L_{cl} = d_m(1 + |\varepsilon_m/\varepsilon_d|) \). Note that even for nanoscale (∼10-nm) metal films, both LRP and SRPs are typically extended to quite a macroscopic scale. The modal indices of SRPs and LRP s in fused silica \((\varepsilon_m = 1.444^2 = 2.085) \) (Ref. 9) gold \((\varepsilon_m = -114.5 + 11.01i) \) (Ref. 10) system at \( \lambda = 1550 \) nm as functions of metal thickness are shown in the first column of Fig. 1.

The splitting of modal indices resulting from the coupling is an electromagnetic analog of a universal phenomenon appearing in linear wave-dynamic systems: energy-level splitting in quantum mechanics, characteristic frequency splitting in acoustics, splitting of relaxation time in coupled ac circuits, etc. Modes in a metal-dielectric multilayer can be represented as a linear combination of surface modes at individual interfaces, with splitting between modal indices determined by the overlapping of the original wave functions.

The system of two metal films supports four modes, which differ by symmetry. Two of these modes are dominated by SPPs propagating at interfaces with the claddings, and their properties are identical to SRPs and LRP s. The other two modes are primarily composed of SPPs propagating inside the gap between the metal films. The mode with antisymmetric magnetic field distribution experiences a cutoff when the gap thickness becomes less than \( d_c = \lambda/2\sqrt{\varepsilon_d} \) and therefore it does not exist in nanoscale structures. The mode with symmetric field distribution, the GPP, survives. For a nanoscale dielectric gap of thickness \( t_g \) between infinitely thick metallic claddings, the modal index \( n_{GPP} \) of gap plasmon polaritons,
is inversely proportional to the gap size. Note that the modal index of the gap plasmon polariton greatly exceeds that of the SRP, providing truly nanoscale-mode confinement. Similar modes exist also in a gap between sharp metallic wedges.\textsuperscript{11} Exact dispersion equations for modes in a gap between metallic claddings were proposed earlier.\textsuperscript{12}

As more layers are added to the structure, the collective interaction of SPPs leads to the mutual repulsion of modal indices. The gap plasmon polaritons contribute to the emergence of high-index supermodes strongly confined to the bulk of the layered material. Since the bulk modes originate from repulsion of GPPs, the total number of these modes is equal to the number of dielectric layers. The entire multilayer may also support surface modes at the interfaces with the claddings. In the case of symmetric claddings they are similar to the LRPCs and SPPCs of a metallic film.

The surface modes behave fundamentally different when \(|\varepsilon_m| < \varepsilon_d|\). Although a single interface does not support a SPP, a thin film of negative-permittivity medium supports two modes with identical field distributions (TM\(_0\)) but different “handedness”—the relation between the directions of the electric and magnetic fields and wave vector of the mode.\textsuperscript{2,13} (Fig. 1, right). One of these modes is a direct “right-handed” analog of a LRP. The other one is its negative-index “left-handed” counterpart. While the analogy is not complete, the negative-index medium is often called optical antimatter.\textsuperscript{14} The close-to-cutoff TM\(_0\)-mode doublet can therefore be considered as an optical mode-antimode pair. A further analogy is seen in the formation of an antisymmetric left-handed gap plasmon polariton in a dielectric gap between metallic claddings when \(|\varepsilon_m| < \varepsilon_d|\).\textsuperscript{2,15,16} Similar to the case of \(|\varepsilon_m| > \varepsilon_d|\), the absolute value of the modal index for the left-handed waves increases as the metal interfaces get closer. Naturally, such an increase of the absolute value of the modal index is accompanied by stronger mode confinement.

Figures 2 and 3 further summarize the modal indices of metal-dielectric composites with 25-nm layers and dielectric claddings, realized in our experiment. While optical losses of the bulk plasmon polariton modes in multilayers are larger than losses of SPPs, these modes are of significant interest to nanophotonics due to extremely strong field confinement. Besides light guiding by subwavelength structures, nanoscale multilayers with appropriately patterned films are promising candidates for the development of metamaterials with negative refractive index.\textsuperscript{16–21} as well as for the development of nanosensors.

The traditional classification scheme for transverse electromagnetic modes in multilayers relies on the number of nodes in the field distributions. Accordingly, the SPP supported by a single interface is a TM\(_0\) mode. The LRP and SRP supported by a thin metal film are labeled as TM\(_0\) and TM\(_1\) modes. The gap plasmon polariton is a TM\(_0\) mode. Such a mode labeling scheme is perfect for dielectric waveguides but causes some confusion when applied to nanoscale metal-dielectric structures. It is a bit inconvenient that the same label TM\(_0\) is designated to rather different electromagnetic excitations such as surface plasmon polaritons, long-range film plasmon polaritons, and gap plasmon polaritons. Two modes of a metal strip with \(|\varepsilon_m| < \varepsilon_d|\) are both TM\(_0\) waves. A further inconvenience is that essentially the same mode—the gap plasmon polariton—is labeled TM\(_0\) in the structure with metallic claddings, and it becomes a TM\(_2\)

\[ n_{GPP} = \sqrt{\varepsilon_d + \frac{1}{2} \left( \frac{\varepsilon_d}{\varepsilon_m} \right)^2} + \sqrt{\left( \frac{\varepsilon_d}{\varepsilon_m} \right)^2 \left( \varepsilon_d - \varepsilon_m \right) + \frac{1}{4} \frac{\varepsilon_d}{\varepsilon_m}} \]
mode in a structure with dielectric claddings, two thin metallic layers, and a guiding dielectric layer. The TMn label indicates the number of nodes in the modal field distribution. This number, however, is not associated directly with the bulk plasmon polariton mode BPP0. In the traditional numerically smooth profile is reasonable to call the fundamental particular interfaces will be more appropriate.

Once the highly confined bulk modes and the film-plasmon-polariton-type modes have distinct properties, it is reasonable to label them differently. In particular, in a structure with a large number of layers, the bulk mode with relatively smooth profile is reasonable to call the fundamental bulk plasmon polariton mode BPP0. In the traditional numbering scheme this is a TMn mode because it has two nodes close to the interfaces with the claddings. Accordingly, the bulk mode of order n (BPPn) would be labeled as TMn+2 in the traditional classification. The labels such as LRP and SRP should be reserved for the modes with intensity maxima at the interfaces with the claddings. When claddings have different permittivities, the labels LRP and SRP become rather senseless. Instead, titles such as SPP bounded to particular interfaces will be more appropriate.

We stress that in a finite-thickness nanolayered film the bulk modes, due to minor penetration into the claddings, are rather independent of the cladding indices. Therefore, all BPPn modes labeling is also independent of the claddings—a significant advantage over the traditional scheme, which is strongly affected by the cladding indices.

The proposed scheme is illustrated in Fig. 2 (right) in the example of modes of a composite with Nd=4 dielectric and Nm=5 metallic 25-nm-thick layers. The structure supports the long-range plasmon polariton (LRP=TM0), the short-range plasmon polariton (SRP=TM1), and four bulk plasmon polariton modes (BPP0, . . . , BPP3). Both LRP and SRPs show a large penetration into the claddings. In contrast, the bulk plasmon polariton modes are confined within the multilayer with a minor fraction of optical power propagating in the claddings. The fundamental bulk mode BPP0 has a rather smooth field profile. For the highest-order bulk mode, the modal field reveals fast oscillations so that it has opposite signs in neighboring dielectric layers. Note that the modal indices of the BPP modes can be several times higher than the refractive index of the dielectric in the multilayer. The origin of this surprising behavior is in the coupling-induced repulsion of the modal indices of individual gap plasmon polaritons discussed above.

The modal indices of guided modes in a multilayer with alternating 25-nm-thick layers of gold and silica are shown in Fig. 3. As predicted, the number of bulk modes is equal to the number of dielectric layers and the maximal modal index increases with the number of layers increasing. For any given number of layers, modes of higher order have larger losses and larger modal indices. Assuming the number of layers is approaching infinity, N→∞, we find the dispersion relation for the highest- and lowest-order bulk plasmon polariton modes (BPPN and BPP0) by setting periodical boundary conditions and requiring that the magnetic field strength have a node in the middle of every metal layer (BPPN) or not have such a node (BPP0):

\[
\text{tanh} \left( \frac{\pi t_m}{\lambda} \sqrt{\frac{n_{BPP}^2 - \varepsilon_m}{n_{BPP}^2}} \right) \text{tanh} \left( \frac{\pi t_d}{\lambda} \sqrt{\frac{n_{BPP}^2 - \varepsilon_d}{n_{BPP}^2}} \right) + \frac{\varepsilon_d}{\varepsilon_m} \sqrt{\frac{n_{BPP}^2 - \varepsilon_m}{n_{BPP}^2 - \varepsilon_d}} = 0, \tag{2a}
\]

\[
\text{tanh} \left( \frac{\pi t_d}{\lambda} \sqrt{\frac{n_{BPP}^2 - \varepsilon_d}{n_{BPP}^2}} \right) \text{tanh} \left( \frac{\pi t_m}{\lambda} \sqrt{\frac{n_{BPP}^2 - \varepsilon_m}{n_{BPP}^2}} \right) + \frac{\varepsilon_d}{\varepsilon_m} \sqrt{\frac{n_{BPP}^2 - \varepsilon_d}{n_{BPP}^2 - \varepsilon_m}} = 0. \tag{2b}
\]

Equations (2a) and (2b) do not contain the cladding indices or the overall composite thickness, further indicating the “bulk” origin of these modes. In the limit of thin (nanoscale) layers, Eqs. (2a) and (2b) yield the following approximation for the modal indices:

\[
\frac{1}{n_{BPP}^2} \approx \frac{1}{n_{BPP}^2} - \frac{k^2}{\pi^2} \text{Im} \varepsilon_m, \quad \frac{1}{n_{BPP0}^2} \approx \frac{1}{n_{BPP0}^2} + \frac{k^2}{\pi^2} \text{Im} \varepsilon_m + \text{Im} \varepsilon_d. \tag{3}
\]

Note that the wavelength disappears from the expression for the modal index of the fundamental mode and the equation becomes equivalent to the predictions of the effective medium theory. The highest-order mode has a larger modal index than a single-gap plasmon polariton provided that t_d,m << λ/π|\varepsilon_m|. A particular case of the fundamental mode, the one with the smallest losses, was studied in the 1950s. SPPs supported by a structure consisting of few metal and dielectric thin films were studied earlier. Nonlocal corrections to the averaged permittivity in metal-dielectric nanocomposites are reported in our recent paper. A similar effect has been found in GHz systems with conducting wire-shaped inclusions.

For the gold-silica multilayers with t_d=t_m=25 nm, Eq. (2) gives Re(\text{Im} n_{BPP})=3.1488 and \alpha=(4\pi/\lambda)\text{Im}(\text{Im} n_{BPP})=8959 cm^{-1}—maximal possible values for the modal index and losses for the high-order bulk mode BPPN. Corresponding values for the fundamental mode are Re(\text{Im} n_{BPP})=2.0128 and \alpha=(4\pi/\lambda)\text{Im}(\text{Im} n_{BPP})=462.8 cm^{-1}. These limits are shown in Fig. 3 by horizontal dashed lines.
The modes in a nanoscale metal-dielectric multilayer are relatively easy to predict and simulate numerically, but their experimental verification is challenging due to the deep subwavelength confinement and very high optical losses. In this paper we report experimental studies of guided modes in nanoscale metal-dielectric multilayers. To excite the high-index modes, we used the evanescent light-coupling scheme. High-index material (silicon, \( n_{Si} = 3.48 \)) was used to match the wave vector of light in free space to the wave vector of a guided mode. To access a wider range of modal indices, semicylinder geometry instead of more traditional prism was used. In a similar manner, a high-index semispherical solid immersion lens can be used, but for accurate angular measurement the semicylindrical geometry is preferable. Light from a fiber-coupled tunable (1490–1590 nm) semiconductor laser (Photronics Inc.) was collimated using a 10X objective. The laser was tuned to the wavelength of 1550 nm, which was verified by an optical spectrum analyzer (Hewlett-Packard HP70951B). The spectral width of the laser radiation was below the resolution of the spectrometer (<0.1 nm). The angular reflection spectrum was measured, and data were plotted as a function of the product \( n_Si \sin(\theta) \), where \( \theta \) is the incident angle. In this scale, the intensity minima directly indicate the modal indices of the guided modes excited through the evanescent coupling.

The multilayer structure was designed to consist of three pairs of silica (~25 nm)/gold (~25 nm). The layers were deposited directly on the flat facet of the semicylinder. The gold layers were deposited by electron beam evaporation and silica layers by plasma-enhanced chemical vapor deposition. With two dielectric gaps between three metal layers, the structure supports two BPP modes. The thickness of the layers was chosen to ensure that the effective indices of the bulk modes are comfortably in the measurement range (~3.0). The first silica layer between silicon and gold is not crucial. Its role is to adjust the evanescent coupling strength for clear observation of guided modes. The experimental data are shown in Fig. 4 on the left. They verify the guided modes with modal indices 2.31 and 2.88 recognized as BPP0 and BPP1. The structure was designed for measuring the modal indices of the bulk modes, while the SPP at the interface with the substrate is overdamped and the SPP at the interface with air has a vanishing small evanescent coupling with the incident beam. By fitting the experimental data with numerically simulated angular reflection, the best-fit structure has been identified as Si/(33 nm silica)/(24 nm gold)/(24 nm silica)/(26 nm gold)/(31 nm silica)/(24 nm gold) [on average, (29 nm silica)/(25 nm gold)], which is close to the deposition target numbers. Figure 4 also shows the modes’ profiles in the manufactured structure calculated using the best-fit data for the layers’ thicknesses.

In conclusion, we analyzed TM-polarized modes supported by nanoscale metal-dielectric multilayers. We show that, in addition to the long-range and short-range plasmons supported by thin films, there are high-index guided modes strongly confined within the bulk of the multilayer. The dispersion relation is derived for the highest-order and lowest-order modes in approximation of an infinite number of layers. A classification scheme is proposed for the modes supported in nanoscale metal-dielectric multilayers. High-index highly confined modes in a structure made of three pairs of gold (~25 nm)/silica (~29 nm) layers are experimentally verified.

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