

Arrays of nano-sized metal structures can be manipulated to create plasmon waveguides that can be smaller than a wavelength.

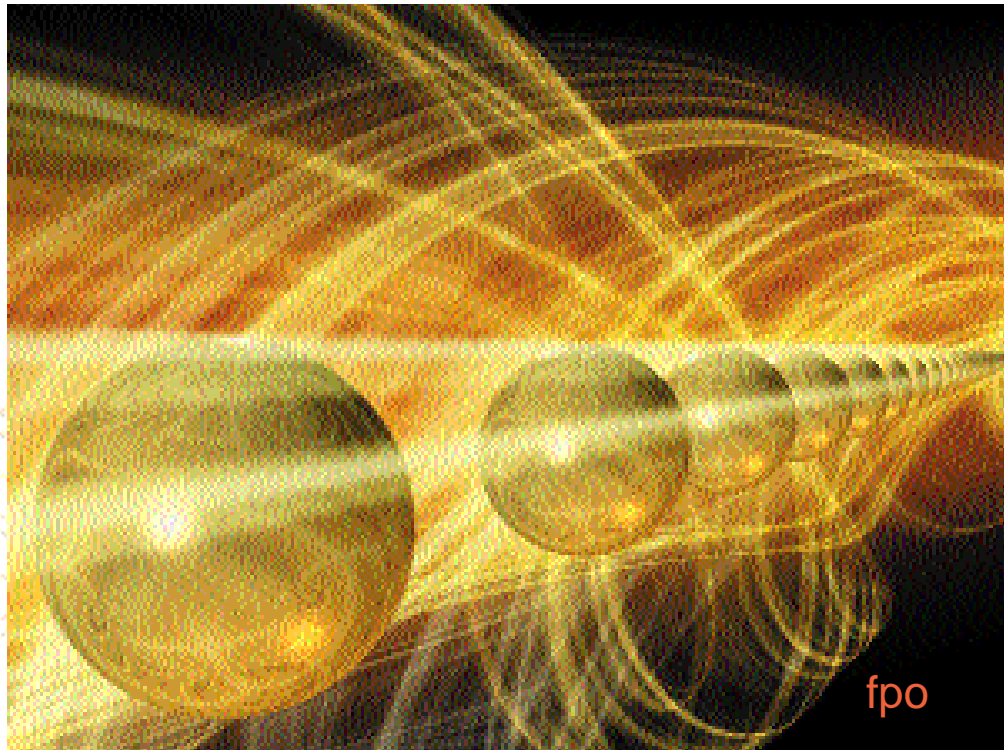


ILLUSTRATION BY DON BISHOP

GUIDING LIGHT

By Harry Atwater, California Institute of Technology

We are entering an age of integrated optics and optoelectronic devices, but few researchers have considered what the ultimate physical limits of device integration might be. The size and density of an integrated photonic device would seem to be controlled by the diffraction limit of light. For a guided mode in a conventional waveguide, this diffraction limit holds the minimum size to be on the order of at least $1/2\lambda$; for typical dielectric materials, this amounts to a few hundred nanometers. Conventional planar waveguide devices are also limited in their geometry and layout by the need to avoid sharp bends that incur large radiative losses.

The unique characteristics of photonic-crystal structures overcome the bend problem, guiding light through sharp 90° corners, but these structures are also limited in critical dimensions and packing density. In order to confine light, the photonic-crystal material requires a periodic structure, with the period size comparable to a few wavelengths.

There may be another method to make optical devices that are considerably smaller than the wavelength of the light being propagated while enabling strong localization, guiding around sharp corners, and switching of light at dimensions below the diffraction limit. Appropriately designed metallic and metallodielectric nanostructures can strongly localize and

manipulate light. In particular, interesting phenomena occur in nanoscale structures at the plasmon frequency, at which optical absorption is resonantly enhanced. [WE NEED TO DEFINE WHAT A PLASMON IS, E.G. "A plasmon is a collective excitation for quantized oscillations of the electrons in a metal, and the plasmon frequency is XXX.]

Periodic arrays of metallic structures embedded in dielectric media might be able to guide and modulate light transmission in a regime dominated by near-field coupling.¹ Our group has coined the name plasmon waveguides for structures operating on this principle and the name plasmonics for the field of study to draw attention to the energy guiding mechanism via surface plasmons.² We have also demonstrated guiding properties of periodic metal structures in a macroscopic analog of plasmon waveguides operating in the microwave regime, thus confirming that energy is coherently transported in these subwavelength guiding structures with the electric field laterally confined to dimensions on the order $\lambda/20$.³ Energy propagation through corners and tee structures is possible. One can even design an all-optical switch using these subwavelength structures.

nanoscale metal structures

Metal structures, with their high reflection and absorption coefficients, are seldom considered as optical waveguides. At the

nanoscale, however, even metallic components become semitransparent. Researchers are investigating metallic nanostructures that guide light for applications, including ultra-compact optical functional devices, light-harvesting elements for molecular and nanocrystalline-based photovoltaic devices, lithographic patterning at deep subwavelength dimensions, and aberration-free lenses that enable optical imaging with unprecedented resolution.

The structures of interest for plasmonics are small particles, wires, rods, and thin films of metals that act as dipole oscillators and whose plasmon frequencies are in or near the visible regime, with plasmon lines incurring minimal interference from interband optical transitions. We have concentrated our research on structures composed of arrays of nanoparticles and films of silver and gold.

For gold and silver particles smaller than 50 nm, the electric dipole moment dominates higher-order optical multipolar moments, and absorption exceeds scattering as a mechanism for optical extinction.⁴ The plasmon linewidth is determined by electron-phonon relaxation ($\tau = 4$ fs for gold and $\tau = 10$ fs for silver). For silver, the plasmon peak dominates the optical properties from 300 to 2000 nm. For gold, interband transitions also contribute at short wavelengths.

For very small particles less than 10 nm, enhanced scattering from the particle surface broadens the plasmon linewidth, so we have concentrated on structure sizes between 10 and 50 nm. Small particles in this regime exhibit a resonant enhancement at the Fröhlich frequency ω_F where $\epsilon_p = -2\epsilon_m$ with ϵ_p and ϵ_m defined as the real parts of dielectric function for the particle and host matrix, respectively. The near-field contribution to the optical scattering cross-section also dominates the far-field contribution; for 30-nm gold particles, the near-field cross-section is one order of magnitude higher than the far-field

scattering cross-section and is size independent to the first order.

plasmon waveguides

The strong interaction of individual metal nanostructures⁵ with light can be used to fabricate waveguides if energy can be transferred between the structures. We showed that the dipole field resulting from a plasmon oscillation in single metal nanoparticles can induce a plasmon oscillation in a closely spaced neighboring particle due to near-field electrodynamic interactions. J. Krenn's group [AFFILIATION TO COME] showed that ordered arrays of closely spaced noble metal particles show a collective behavior under broad beam illumination.⁶ Their finding supports such an interaction scheme.

When metal nanostructures are separated by only a few tens of nanometers, the electric dipole interaction is dominated by the near-field term, which is strongly dependent on distance. The interaction strength and the relative phase of the electric field in neighboring particles both depend on polarization and frequency. This interaction leads to coherent modes with a wave vector k along the nanoparticle array.

One can calculate a dispersion relation for energy propagation along the nanoparticle chain by taking into account n^{th} neighbor interactions via a polarization-dependent interaction frequency ω_1 derived from the electromagnetic interaction term and the plasmon dipole resonance ω_0 . The model also accounts for internal and radiative damping. Calculations for modes with the electric field polarized both parallel and perpendicular to a linear array of metal nanoparticles show the interaction dominated by nearest neighbor coupling.

For both polarizations, the propagation velocity of the guided energy, given by the slope $d\omega/dk$ of the dispersion relation, is highest at the resonance frequency ω_0 . Calculations for 50-nm silver spheres with a center-to-center distance of 75 nm show

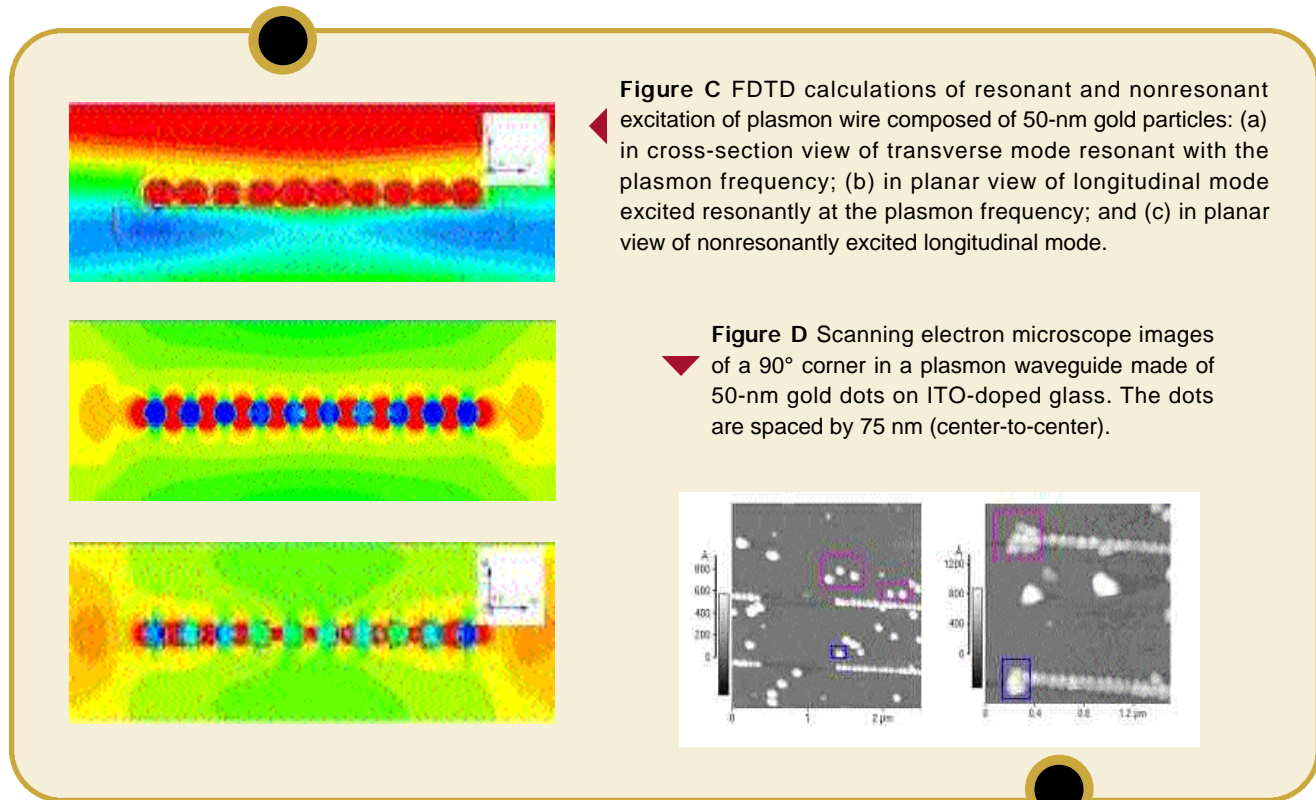


Figure C FDTD calculations of resonant and nonresonant excitation of plasmon wire composed of 50-nm gold particles: (a) in cross-section view of transverse mode resonant with the plasmon frequency; (b) in planar view of longitudinal mode excited resonantly at the plasmon frequency; and (c) in planar view of nonresonantly excited longitudinal mode.

Figure D Scanning electron microscope images of a 90° corner in a plasmon waveguide made of 50-nm gold dots on ITO-doped glass. The dots are spaced by 75 nm (center-to-center).

energy propagation velocities of about 10% of the speed of light. This is several times faster than the saturation velocity of electrons in typical semiconductor devices.

What about loss from the waveguide? Plasmon waveguides can suffer losses caused by both radiation into the far field and internal damping. We expect radiative losses into the far field to be negligible due to the dominance of near-field coupling. Internal damping of the surface plasmon mode results from resistive heating and was shown to induce transmission losses of about 6 dB/ μm . While this is a high loss per length, the possibility of creating an entire device smaller than 1 μm^2 implies that the loss per function may be comparable with other integrated photonic devices.

We have also analyzed the transmission characteristics of several circuit elements such as corners and tee structures. Due to the near-field nature of the coupling, signals can be guided around 90° corners and split via tee structures without radiative losses into the far field at the discontinuity. We calculated power transmission coefficients for guiding energy around corners and for signal splitting in tee structures by requiring continuity of the plasmon amplitude and of the energy flux at the corner where the wave is partly transmitted and reflected. The transmission coefficients are a strong function of the frequency of the guided wave and of its polarization and show a maximum at the dipole plasmon frequency. Transmission coefficients close to 100% are possible for propagation around 90° corners for certain polarizations, and we calculated lossless signal splitting in tee structures. Because the light being guided is coherent, such structures could be used to design switches that use interference effects, such as Mach-Zehnder interferometers.

We can see energy localization at the plasmon frequency using 3-D finite difference time domain (FDTD) calculations. We calculated energy localization for a plasmon wire of 50-nm gold particles with $k=0$ modes excited in resonance with the plasmon frequency (see figure C on page 43). These simulations indicate energy localization to dimensions of approximately 10% of the exciting wavelength of 520 nm.

fabrication and characterization

Because position and width of the dipole resonance depend on the shape and size of the metal particle, we need a fabrication approach that produces particles with a narrow size distribution. Furthermore, particles must be placed with regular spacing because the transport properties depend strongly on the distance between them.

Our initial structures use gold nanoparticles with diameters between 30 and 50 nm as building blocks for plasmon waveguides. Gold particles in this size regime are small enough to allow for efficient excitation of the surface dipole plasmon mode only and large enough that they do not suffer from enhanced damping due to surface scattering of the conduction electrons. Because the gold plasmon resonance in these particles is conveniently located around 514 nm, we can excite them with 514-nm emission from an argon laser.

We use electron-beam lithography and atomic-force microscope (AFM) techniques to fabricate plasmon waveguides. Electron-beam lithography provides excellent size and distance control of the nanoparticles constituting the waveguides (see figure D on page 43). Using electron-beam lithography, structures can be fabricated using any material that withstands

the liftoff process, such as metals and various oxides.

We manipulated randomly deposited nanoparticles with the tip of an AFM into a straight line using a dedicated AFM system and software developed by our colleagues Ari Requicha and Bruce Koel at the University of Southern California (Los Angeles, CA). We chose the center-to-center distance between particles to be three times the particle radius in order to optimize the guiding properties of the structures. Our control of the particle spacing is limited by the spatial resolution of the microscope. AFM manipulation is compatible with complex materials such as core-shell particles. Polymer beads containing dye molecules can be precisely manipulated into position using the technique.

We confirmed experimentally the existence of coupled dipole plasmon modes of plasmon wires using far-field ($k=0$) extinction measurements.⁷ Extinction spectra for gold plasmon wires taken for longitudinal and transverse polarizations show distinct shifts in the plasmon frequency and exhibit a dependence on interparticle spacing in excellent agreement with the plasmon wire theory described above. For resonant wire excitation at 590 nm ($\omega_0 = 3.19 \times 10^{15}$ rad/s), which corresponds to the frequency of maximum group velocity, the measurements indicate a longitudinal mode group velocity of 4.0×10^6 m/s and a bandwidth of the dispersion relation of 26 nm (1.4×10^{14} rad/s). These numbers indicate a group velocity for these structures of approximately 1% of the speed of light in free space.

We are focusing our efforts on observation of modes with $k > 0$ by using a near-field scanning optical microscope as a spatially localized excitation source for plasmon wires. In this way we propose to characterize power transmission by exciting one end of a plasmon wire and detecting far-field emission from fluorescent probes at the other end.^{8,9}

A near-field scanning optical microscope enables simultaneous near-field imaging and noncontact AFM imaging. Thus, we can correlate light scattering, guiding, and loss with the physical microstructure of the nanoparticles array. This general scheme will enable us to experimentally characterize power transmission in plasmon waveguides and validate the dipole approximation models and FDTD simulations. **oe**

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