ADVANCED MATERIALS

Plasmonics—A Route to Nanoscale Optical Devices**

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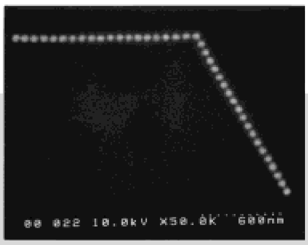

The further integration of optical devices will require the fabrication of waveguides for electromagnetic energy below the diffraction limit of light. We investigate the possibility of using arrays of closely spaced metal nanoparticles for this purpose. Coupling between adjacent particles sets up coupled plasmon modes that give rise to coherent propagation of energy along the array. A point dipole analysis predicts group velocities of energy transport that exceed 0.1c along straight arrays and shows that energy transmission and switching through chain networks such as corners (see Figure) and tee structures is possible at high efficiencies. Radiation losses into the far field are expected to be negligible due to the near-field nature of the coupling, and resistive heating leads to transmission losses of about 6 dB/μm for gold and silver particles. We analyze macroscopic analogues operating in the microwave regime consisting of closely spaced metal rods by experiments and full field electrodynamic simulations. The guiding structures show a high confinement of the electromagnetic energy and allow for highly variable geometries and switching. Also, we have fabricated gold nanoparticle arrays using electron beam lithography and atomic force microscopy manipulation. These plasmon waveguides and switches could be the smallest devices with optical functionality.

1. Introduction

In recent years, there has been tremendous progress in the miniaturization of optical devices. Planar waveguides and photonic crystals are currently key technologies enabling a revolution in integrated optical components.[1,2] The size and density of optical devices employing these technologies is nonetheless limited by the diffraction limit of light, which imposes a lower size limit on the guided light mode of about $\lambda/2n$ (a few 100 nm).[1] Another limitation is the typical guiding geometry. Whereas photonic crystals allow for guiding geometries such as 90° corners,[2] planar waveguides are limited in their geometry because of radiation leakage at sharp bends.[1] Scaling optical devices down to the ultimate limits for the fabrication of highly integrated nanophotonic devices and circuits requires electromagnetic energy to be guided on a scale below the diffraction limit and that information can be guided around 90° corners (bending radius $\ll$ wavelength of light).

Recently, a new method for the guiding of electromagnetic energy has been proposed that allows for a further reduction of the device size to below the diffraction limit in a variety of geometries.[3,4] It was shown that electromagnetic energy can be guided in a coherent fashion via arrays of closely spaced metal nanoparticles due to near-field coupling. After this initial paper, a number of publications discussing similar structures appeared.[5,6]

In this paper, we will describe the findings of our theoretical and experimental work on metal nanoparticle waveguides, and their macroscopic microwave frequency analogues. In Section 2, we report on theoretical modeling of nanoparticle waveguides and show dispersion relations of propagating modes in such waveguides. In Section 3, we present experimental verification of the guiding properties of periodic metal structures in a macroscopic analogue of nanoparticle waveguides, operating in the microwave regime.[5,6] Our results confirm that energy is coherently transported in these subwavelength guiding structures and that energy propagation through corners and tee structures is possible and that an all-optical switch can be designed. Additionally, the electric

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field turns out to be laterally confined to dimensions of the order $\lambda/20$. Finally, in Section 4 we discuss different approaches to the fabrication of nanoparticle waveguides.

2. The Key Ingredients of Nanoparticle Waveguides: Plasmon Resonance and Near Field Coupling

Individual noble metal nanoparticles strongly interact with visible light at their dipole surface plasmon frequency due to the excitation of a collective electron motion (a so called plasmon) inside the metal particle.[7] The surface of the nanoparticle confines the conduction electrons inside the particle and sets up an effective restoring force leading to resonant behavior at the dipole surface plasmon frequency. Figure 1 shows the flow of electromagnetic energy around a single spherical metal nanoparticle at two different excitation frequencies. When the frequency of the light is far from the intrinsic plasmon resonance of the metal nanoparticle (Fig. 1a), the energy flow is only slightly perturbed. At the plasmon resonance frequency, the strong polarization of the particle effectively draws energy into the particle (Fig. 1b). This effect can be observed as a strong enhancement in the scattering cross section in optical extinction measurements.[7] The dipole surface plasmon resonance is most pronounced for particles much smaller than the wavelength of the exciting light, since in this case all conduction electrons of the particle are excited in phase. The resonance frequency is determined by the particle material, the shape of the particle and by the refractive index of the surrounding host. Surface plasmons can be efficiently excited in the noble metals gold, silver and copper due to their free electron like behavior. For these metals the plasmon resonance occurs in the visible range of light in a variety of hosts.

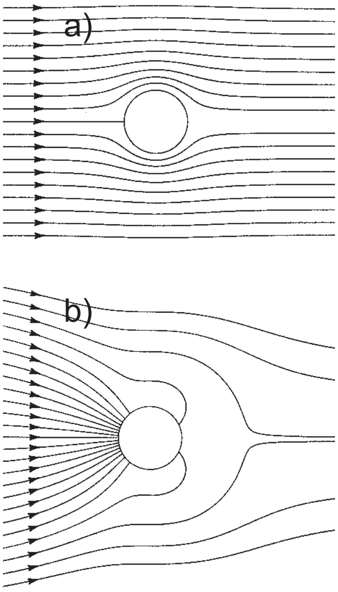

Fig. 1. Energy flux (Poynting vector) around a metal nanoparticle under plane wave excitation at two frequencies: a) When the excitation occurs far from the plasmon resonance frequency, the energy flow is only slightly pertubed. b) When the excitation occurs at the plasmon frequency, the energy flow is directed towards the particle. This resonant field enhancement is a key element of plasmon waveguides. Image taken from C. F. Bohren, D. R. Huffman, Absorption and Scattering of Light by Small Particles, copyright Wiley, New York 1983; this material is used by permission of John Wiley & Sons, Inc.

The strong interaction of individual metal nanoparticles with light can be used to fabricate waveguides if energy can be transferred between nanoparticles. In fact, we have shown[3] that the dipole field resulting from a plasmon oscillation in a single metal nanoparticle can induce a plasmon oscillation in a closely spaced neighboring particle due to near-field electrodynamic interactions. The recent finding that ordered arrays of closely spaced noble metal particles show a collective behavior under broad beam illumination supports such an interaction scheme.[8] We propose 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.

In the following, we will discuss the nature of the propagating modes. When metal nanoparticles are spaced closely together (separation a few tens of nanometers), as depicted in the inset of Figure 2, the strongly distance dependent near field term in the expansion of the electric dipole interaction dominates. The interaction strength and the relative phase of the electric field in neighboring particles are both polarization and frequency dependent. This interaction leads to coherent modes with a wavevector k along the nanoparticle array. One can calculate a dispersion relation for energy propagation along the nanoparticle chain by taking into account nth neighbor interactions via a polarization dependent interaction strength ω_1 derived from the electromagnetic interaction term and the plasmon dipole resonance ω_0. Internal and radiation damping are also accounted for in the model.[3] Figure 2 shows the results of such a calculation for modes with the electric field polarized along the chain (longitudinal modes L)

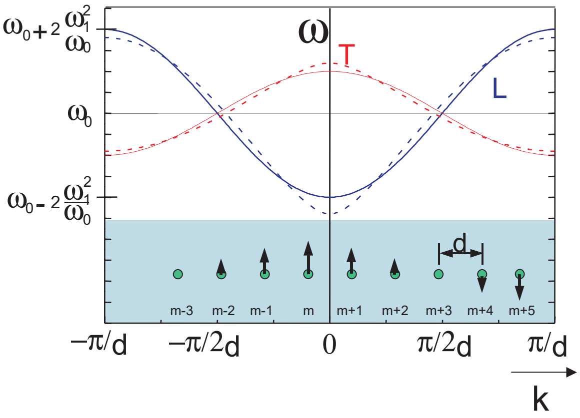

Fig. 2. Dispersion relation for plasmon modes in a linear chain (see inset) of metal nanoparticles, showing the twofold degenerate branch corresponding to transverse modes (T) and the branch corresponding to longitudinal modes (L). Results are shown for calculations incorporating nearest-neighbor interactions only (solid curve), and including up to fifth-nearest neighbor interactions (dotted curve). The small difference in the dispersion curves illustrates that mode propagation in plasmon waveguides is dominated by near-field interactions.

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and for modes polarized perpendicular to the chain (transverse modes T). Calculations were done including nearest-neighbor coupling only (solid lines) and including up to five nearest neighbors in the coupling term (dotted lines) for an infinite linear array of metal nanoparticles. The inclusion of up to five nearest neighbors has little effect on the dispersion curves, confirming that the interaction is dominated by nearest neighbor coupling. For both polarizations, the propagation velocity of the guided energy, given by the slope $\mathrm{d}\omega/\mathrm{d}k$ 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 energy propagation velocities of about 10 % of the speed of light. This is ten times faster than the saturated velocity of electrons in typical semiconductor devices.

Aside from the dispersion relation, another important parameter in waveguide design is loss. In plasmon waveguides, the loss can be due to radiation into the far field and due to internal damping. Radiation losses into the far field are expected to be negligible due to the dominance of near field coupling. Internal damping of the surface plasmon mode is due to resistive heating. This damping was shown to induce transmission losses of about 6 dB/µm.

We further analyzed the transmission characteristics of several circuit elements such as corners and tee structures (Fig. 3). Due to the near field nature of the coupling, signals can be guided around 90° corners and split via tee structures with negligible radiation losses into the far field at the discontinuity. Power transmission coefficients for the guiding of energy around corners and for signal splitting in tee structures were calculated by requiring continuity of the plasmon amplitude and of the energy flux at the corner where the wave gets 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 lossless signal splitting was shown in tee structures.

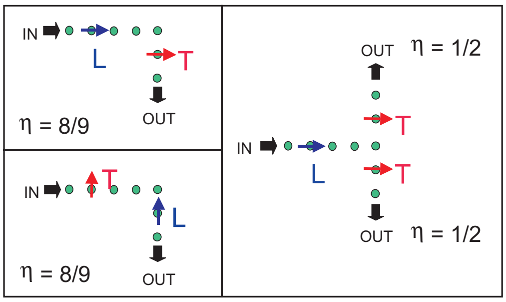

Fig. 3. Calculated power transmission coefficient η in nanoparticle chain arrays for a 90° corner and a tee structure. The black arrows indicate the direction of the energy flow, the blue arrows indicate a longitudinal mode (L), and the red arrows indicate a transverse mode (T). An η value of 1 corresponds to 100 % transmission.

Due to the coherent nature of the propagation, it is also possible to design switches that rely on interference effects, such as Mach–Zehnder interferometers.[3] An example of such a switch will be discussed in Section 3.

3. Macroscopic Analogues to Plasmon Waveguides—Yagi Arrays

In this section, we present large scale analogues to plasmon waveguides and analyze them both via experiment and full-field electromagnetic simulations. These macroscopic structures are built out of closely spaced cm-scale metal rods, akin to Yagi antennas.[9] Such structures can be efficiently excited in the microwave regime as outlined in the literature.[5,6] The main difference between these macroscopic structures and plasmon waveguides is that macroscopic rod arrays cannot be excited resonantly.[5] Nevertheless, efficient coupling can be obtained by choosing a rod height just below $\lambda/2$ as described in the literature.[5] Note that in this size range intermediate field and far field terms also enter in the interaction between the rods. This ultimately leads to radiation losses into the far field at positions where a propagating wave changes direction, as will be shown below. We expect these radiation losses to be negligible in nanoscale plasmon waveguides due to the purely near field nature of the coupling.

For our microwave experiments, we used thin copper rods of 14 mm height to build guiding arrays such as straight lines and corners. The rod spacing was 2 mm. The structures were excited at 8 GHz (λ = 3.7 cm) via a microwave dipole source. To measure propagation loss, we probed the field around the guiding structures using a short probe dipole, which did not influence the field distribution. To confirm our findings, we also conducted full-field electrodynamic simulations to calculate the distribution of the electric field around our structures.

Figure 4 shows simulation results for three different structures. The color field plots depict the absolute value of the *z*-component of the electric field on a logarithmic scale spanning four orders of magnitude. Figure 4a shows the calculated electric field around a tee structure. The energy is highly confined to the guiding structures (90 % within a distance of $\lambda/20$ from the arrays). Along the straight parts of the tee no energy loss due to radiation into the far field is observed. The ratio of the power in the each sidearm of the tee to the power in the stem is about 8 dB. Also shown is the power variation at a distance of 0.15λ from the array as determined by experiment (black line) using the probe dipole and as calculated (red line). Simulation and experiment are seen to be in good agreement. Information transport in these microwave structures occurs at a group velocity of $0.6c$, and the attenuation along linear arrays is found to be 3 dB/8 cm. At discontinuities where the traveling wave changes direction, e.g., at the junction of the tee structure, the wave gets partly reflected and partly transmitted. The reflected wave establishes a standing wave pattern along the structure, as can be seen in Figure 4. Note that radiation losses at discontinuities are predicted to be strongly reduced in nanoscale plasmon waveguides, due to stronger near field interactions.

Figures 4b and c show an example of an electromagnetic switch based on interference between two sources at positions A and B on two arms of a tee structure. The field strength at

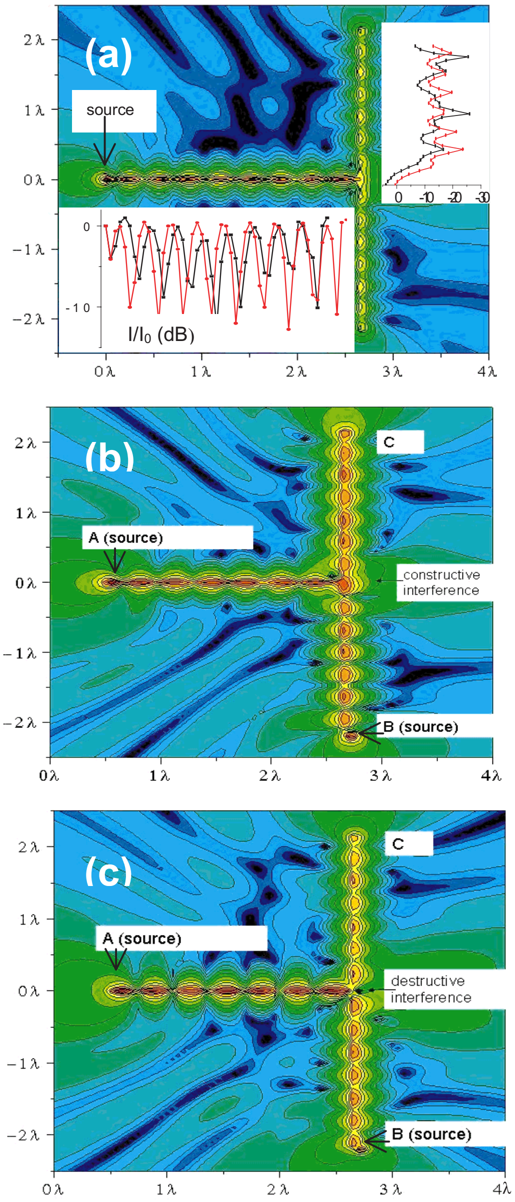

Fig. 4. Calculations and experimental data on large scale analogues to plasmon waveguides, operating in the microwave regime. The guiding structure consists of closely spaced metal rods. The color plots show the distribution of $|E_z|$ in the x–y-plane for a tee structure (a) and a switch (b,c). The color-scale is logarithmic and spans about four orders of magnitude. The inset in (a) shows both the measured (black line) and simulated (red line) power at a distance of 0.15λ from the tee structure. b) In-phase excitation of inputs A and B leads to constructive interference at the junction and an enhanced signal at output C (ON-state). c) Out-of-phase excitation of inputs A and B results in destructive interference at the junction and a reduced signal at output C (OFF-state).

position C in the third arm is determined by the relative phase of the inputs A and B: If the sources are in phase (Fig. 4b), the waves originating at positions A and B interfere constructively at the tee junction. If they are out of phase (Fig. 4c), they interfere destructively. Constructive interference results in a 50 % higher intensity in arm C (Fig. 4b) compared to the case where the sources interfere destructively (Fig. 4c). This corresponds to an ON and an OFF state of a switch. The reason that the switching efficiency is less than 100 % is due to the fact that the energy propagation is not solely due to coupling between nearest neighbors. As can be seen in Figure 4c, even in the case where the waves interfere destructively at the junction rod, there is still energy leaking into arm C due to coupling between rods which are further apart. Such cross talk effects are expected to be strongly reduced in nanoscale plasmon waveguides.

4. Fabrication of Plasmon Waveguides

Here we present different approaches for fabrication of nanoscale plasmon waveguides with optical functionality. We are aiming at realizing all functional structures discussed in Sections 2 and 3.

Since position and width of the dipole resonance depend on the shape and size of the metal particle, the applied fabrication methods should produce a narrow size distribution of the individual particles. Furthermore, a regular particle spacing is crucial for the transport properties due to the strong distance dependence of the electromagnetic near field.

For our initial structures we have chosen 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 to show no enhanced damping due to surface scattering of the conduction electrons.[7] Additionally, the gold plasmon resonance in these particles is conveniently located around the 514 nm line of an Argon laser for excitation. The center-to-center distance between particles was chosen as $3R$, where R is the particle radius, to optimize the guiding properties of the structures, as suggested in the literature.[4]

Our efforts to build functional plasmon waveguides concentrate around two fabrication techniques. The first method is electron beam lithography (EBL). EBL provides excellent size and distance control of the nanoparticles constituting the waveguides. Using EBL structures can be fabricated using any material that withstands the liftoff process, such as metals and various oxides. Figure 5a shows a 60° corner structure fabricated using this method on indium tin oxide (ITO) doped glass. The individual particles are 50 nm in size. Our second method of fabrication uses manipulation of randomly deposited nanoparticles using the tip of an atomic force microscope (AFM). Figure 5b shows an AFM picture of 30 nm gold particles which were pushed on a straight line using a dedicated AFM system and manipulation software developed at the University of Southern California.[10] In this case, the control of the particle spacing is limited by the spatial resolution of the AFM. The advantage of AFM manipulation lies in the fact that complex materials such as core–shell particles can be used.

We are currently investigating the guiding properties of these plasmon waveguides.

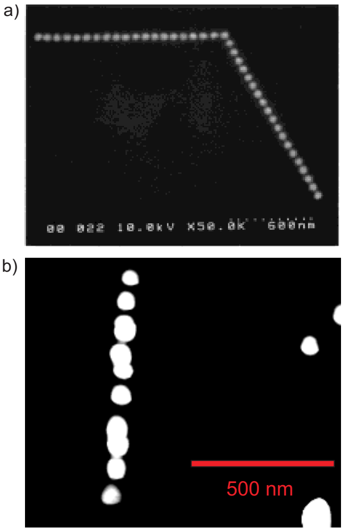

Fig. 5. a) Scanning electron microscopy image of a 60° corner in a plasmon waveguide, fabricated using electron beam lithography. The gold dots are ~50 nm in diameter and spaced by ~75 nm (center-to-center). b) Straight plasmon waveguide made using 30 nm diameter colloidal Au nanoparticles. The particles were assembled on a straight line using an atomic force microscope in contact mode, and subsequently imaged in non-contact mode.

5. Summary

Plasmon waveguides can be used to build nanoscale optical devices with a lateral mode size well below the diffraction limit. Calculations show that basic circuit elements such as corners, highly efficient splitters and switches can be realized. The energy transport can take place at group velocities as high as 10 % of the velocity of light. Experiments on macroscopic analogues to these structures in the microwave regime confirm the guiding mechanism and show efficient guiding around corners and low-loss splitting in tee structures. Fabrication of nanoscale plasmon waveguides is possible using both e-beam lithography and AFM manipulation. These plasmon waveguide structures are promising candidates for future nanoscale integrated optics.

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