Studies of Light Propagation and Localization in Silver Nano Plasmonic Waveguides

A thesis submitted towards partial fulfilment of

BS-MS Dual Degree Programme

By

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under the guidance of

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Certificate

This is to certify that this thesis entitled "The studies of light propagation and localization in silver nano plasmonic waveguides" submitted towards the partial fulfilment of the BS-MS dual degree programme at the Indian Institute of Science Education and Research Pune represents original research carried out by DANVEER SINGH at Indian Institute of Science Education and Research Pune, under the supervision of Dr. G.V. PAVAN KUMAR during the academic year 2011-2012.

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Abstract

We have studied light propagation and localization in nano plasmonic waveguides. We have chemically synthesized silver nanowires (Ag NW) as nanowaveguides whose dimensions were approximately 15 to 20µm in length and 100 -115nm in diameter. We have investigated end-to-end coupled nanowires that are in the shape of “V” or “L”. We illuminated one end of the coupled nanowire which resulted in surface plasmon polariton propagation. These plasmons further get decoupled at discontinuities leading to emission of photons. In this study we have experimentally observed localization of light at the junction and distal ends of coupled nanowires. We have also studied the intensity modulation by polarization-controlled illumination at different parts of the coupled nanowire. This control mechanism acts as intensity switching in end-to-end coupled nanowires. This study has implications in plasmonic circuitry and nano-optical sensors.

Contents

1 Introduction:

1.1 History of plasmons

In 1950's a new phenomena was observed in the sea of electrons and ions i.e. the oscillations of electron densities in plasma. This phenomena was analysed by various experiments in which high speed electrons (tens of KeV) were bombarded on the plasma and tried to observed what happened to their energies[1, 2, 3, 4]. Experiments shows that there is a pattern in the electron energy loss spectrum which indicates the existance of these oscillations. People have also observed that these oscillations are not only confined to the bulk plasma but is also found at the surface of the metals. This phenomena was predicted by R.H. Ritchie[5] in his experiment of electron energy loss spectrum. They used thin metal film and bombarded it by beam of electrons. Later on a series of experiments were done to characterize these collective excitations, and the quanta of these collective surface oscillations are called surface plasmons.

After completely analyzing plasmonic phenomena, researcher have found a number of applications. The main issue for these applications is the behaviour of plasmons to get coupled with photons forms surface plasmons polaritons (SPP) which propagate along the interface of dielectric and metal surface. With the help of these surface plasmons we can propagate light in sub wavelength dimensions beyond diffraction limit[6]. There is another kind of surface plasmons called localized surface plasmons (LSP) which are confined within the very small space. LSP can analyze molecules at single level with the help of surface enhanced raman spectroscopy (SERS). Reseacher have found major applications of this phenomena in optical circuits[7, 8] where they used plasmons as information carrier, biochemical sensors [9, 10], nano-optical logic gates, plasmonic routers, modulaters and switches[11]. Such plasmons can couple with photons and form a polaritons which propagate along the surface of this structure. Propagation is very sensitive to the rouhgness of the surface where SPPs are propagating and hence can be used to measure the surface roughness[12], so we can use this technique to transfer the information much faster than the currently existing technology. Structures can generally be nanowires, nanoribbons, chains of nanoparticles, etc. Plasmonic nanostructures can be used as waveguides to serve as nano-optical logic gates, plasmonic routers, modulaters and switches etc.

1.2 Physics behind the surface plasmons

The optical properties of metals can explained by plasma model, where a sea of electrons move against fixed positive ions. When we applied electromagnetic field, electrons will oscillate in response to it and their motion got damped due to the collisions with characteristic collision frequency $\Upsilon = 1/\tau$. τ is known as the relaxation time of free electron, which is approximately 10^{-14}s, so the Υ = 100THz. The equation of motion of an electron is $m\ddot{x} + m\ \Upsilon\dot{x} = -\ eE$. If we assume that the applied electromagnetic field is in the form of harmonic in time dependence, $E(t) = E_o exp(-i\omega t)$, then the solution for the motion of an electron will be in the form

$$X(t) = X_0 exp(-i\omega t).$$

$$X(t) = \frac{eE(t)}{m(\omega^2 - i\Upsilon\omega)}$$

Where

$$X_0 = \frac{e}{m(\omega^2 - i\Upsilon\omega)}$$

So, as we know displaced electron will contribute to the macroscopic polarization

$$P = -nex$$

$$P = \frac{ne\hat{}2E(t)}{m(\omega^2 - i\Upsilon\omega)}$$

As we know displace ment ,D, is given by

$$D = \varepsilon_0 E + P$$

hence

$$D = \varepsilon_0 E(t) - \frac{ne\hat{}2E(t)}{m(\omega^2 - i\Upsilon\omega)}$$

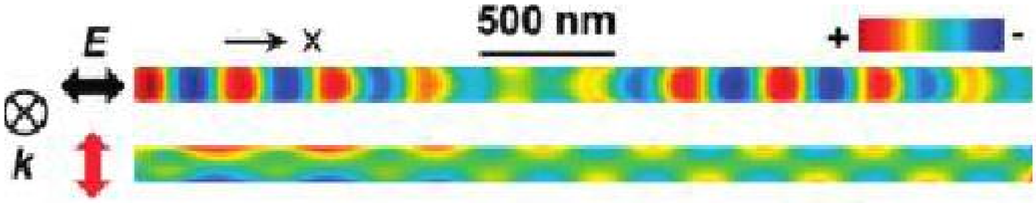

Figure 1: The charge distribution on the surface of wire, arrow shows the direction of polarization, colors red to blue represents positive to negative. k- vector shows direction of incidence and x represents direction of propagation of SPPs .[22]

$$D = \varepsilon_0 E(t)[1 - \frac{\omega^2{}_p}{\omega^2 - i\Upsilon\omega}]$$

where,

$$\omega^2{}_p = \frac{ne\hat{}2}{m\varepsilon_0}$$

This ω_{p} is called frequency of bulk plasmons.

Now we can derive the dielectric function of this medium

i.e

$$\varepsilon(\omega) = 1 - \frac{\omega^2{}_p}{\omega^2 - i\Upsilon\omega}$$

Now, if light is illuminate at the interface of metal and dielectric then the surface wave generate which propagate along this interface with evanescent decay in perpendicular direction to the interface. This waves are bound to the surface and its quanta is called surface plasmon polariton (SPP). The relation between bulk plasmon frequency and surface plasmon frequency is given by :

$$\omega_{sp} = \frac{\omega_p}{\sqrt{1 + \varepsilon_2}}$$

where ε_2 is the dielectric permittivity of dielectric medium[30].

The SPPs propagate at the interface of two medium in the form of oscillating charges in up and down direction as shown in fig.1

If light is illuminated at the surface of metal, then there are two fundamental modes that excites: m=0, m=1. If illuminated light is parallel polarizated then m=0 mode will get excite and if illuminated light which has perpendicular polarization then m=1 will excite. In m=0 mode the charge density is uniform around the azimuth at given position of this nanowire whereas in m=1 mode charge oscillate in transverse direction and hence charge distribution have two nodes around the nanowire. At any arbitrary polarization both modes excited and propagate along the interface. The net charge distribution is superimposition of both these modes. These excitation modes damped over a distance because some losses. These losses can be due to surface roughness , in-coupling efficiency, shape and size of nanostructure [22] etc. The intensity of these modes decreases exponentially which can be calculated by propagation length (L_o) where propagation length is the length at which the intensity of SPPs decreases to 1/e times initial value. So the expression for the intensity of SPPs as a function of distance are given below:

$$I(x) = I_o exp(-x/L_o)$$

hence

$$L_o = x/exp(I_o/I(x))$$

so, the propagation loss (ß) [13] will

$$ß = -10 log(1/e)/L_o$$

Recent studies show that the gold and silver nanowaveguides can propagate SPPs upto tens of micro-meter with low propagation losses [14, 15, 16, 17, 18, 19, 20, 21]. The nanowires can be produced by top-down approach using lithographical methods and bottom-up approach by using chemical process.

2 Methods

2.1 Preparation of nanowires

We prepared two solutions A and B at room temperature, solution A is 0.1M $AgNO_3$ (Sigma Aldrich) in 3ml of ethylene glycol(EG)(Sigma Aldrich) and solution B is 0.6M Poly (Vinyle pyrrolidone) (PVP) , (molecular weigth~ 55000 Sigma Aldrich) in 3ml of EG. These two solutions, A and B, were mixed with each other and 6ml of this solution added to the 5ml of EG which was preheated for one hour at 160 °C in the round bottom flask. The mixture of a solution of A and B was injected by two way channel at the rate of 1ml in 3min to control the aspect ratio and yield . After injecting completely, the solution was heated again at 160 °C for 1h . In the solution obtained had nanowires and nanoparticles. To remove nanoparticles, EG and PVP (surfactant), we centrifuged this diluted solution, 3 times with ethanol to remove EG and PVP and 10 times with water to remove nanoparticles at 2000 rpm to 3000 rpm for 10 min. By this method we obtained variety of nanowires, among them we considered pairs of Ag Nanowires: two Ag nanowire coupled by their ends, as shown in figure1. This chemical process resulted in nanowires 10µm to 30µm long and 100nm to 120nm in diameter nanowires which are well suited for optical studies.

2.2 Preparation of sample for testing light propagation and localization

The solution contained various shapes of configurations of nanowires with negligible amount of nanoparticles. All these nanowires dispersed in the water solvent. The sample was prepared by using dropcast method in which a glass slide was cleaned by detergent and rinsed with deionized water. After that, glass slide was wiped with acetone to remove organic impurities. We took a drop of solution containing Ag NWs and dispersed it gently on the cleaned dried glass slide and dried it at room temperature in dust free environment (we used dessicator for this purpose).

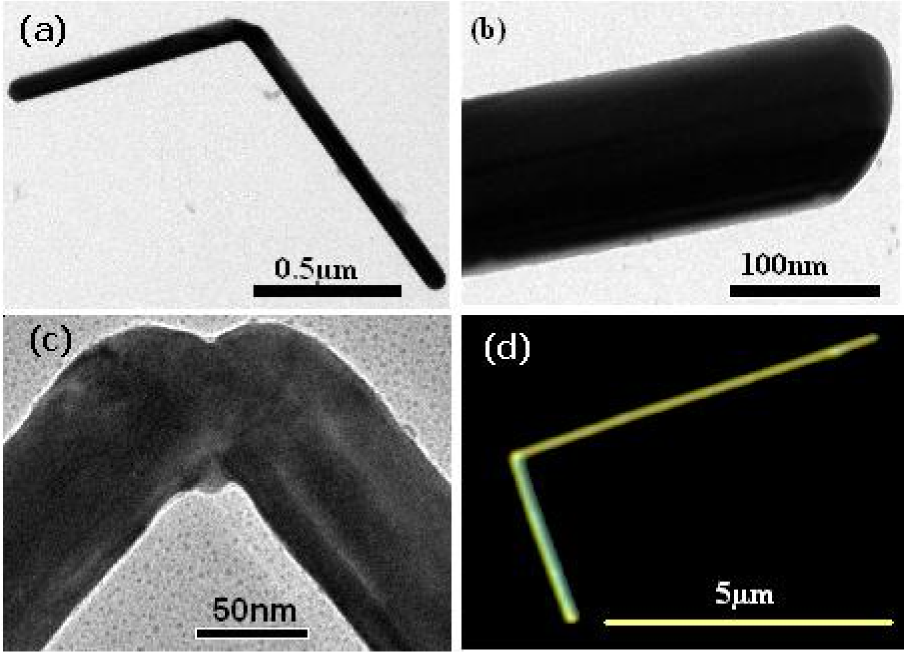

Figure 2: End to end coupled nanowires used in this study.(a) TEM image, (b) TEM image of one end of this wire at higher resolution, (c) Nanowire junction at 50nm scale, (d) The Dark field image of nanowire pair.

3 Results

3.1 Light propagation and localization in end to end coupled AgNW

In this experiment we have studied the plasmon assisted light propagation and localization in end to end coupled silver nanowires. For this experiment we used the upright microscope (Olympus BX – 53) which has both facilities of brightfield and darkfield illumination modules and a CCD camera. For testing the light propagation and localization, the prepared sample was placed, under the high numerical aperture(NA) objective lense(100x, 0.9NA). We have choosen a nanostructure whose configuration is similar to the one shown in fig1(a), 1(d) and 1(e). For testing the light propagation, we took end to end coupled nanowire as shown in figure3(a) where length of wire was 6.6µm + 5.6µm and angle between their two nanowire is 120o. This end to end coupled nanowire was illuminated by tightly focused laser light of wavelength 632.81nm, at one end of NW and captured the optical mages in the presence and absence of brightfield illumination as shown in fig.3(a) and 3(b). In this optical images we observed the illuminated nanostructure emitting light at the junction and at the distal end.

3.1.1 Light propagation capabilities of end to end coupled AgNW

For testing the light propagation capabilities of end to end coupled, AgNW as shown in fig.3(a), illuminated by tightly focused laser beam of 632.81nm wavelength under high numerical aperture (NA) objective lens (100x, 0.9 NA) at one end of this kind of nanostructure and captured the optical images. We observed bright spots at the junction and at the distal end. Fig.3(c) shows the 3-dimensional representation of optical image which is shown in fig.3(b). In fig.3(c) there are three peaks where intensed peak is of illuminated laser light and other peak is at the junction and weakest peak is from the distal end. We calculated the propagation loss of this nanostructure as $-10\log(I/I_o)/L_o$ expressed in dB (decibel), where I is the emitted light intensity from the junction and distal end. We found the propagation loss for wire shown in fig.3(c) is 16.35dB up to juction and 25.07dB up to distal end. This indicates that some of the bound SPPs are decoupled at the junction emitting free space photons.

3.1.2 Light localization capabilities of end to end coupled AgNW

The plasmons localization capability of this nanostructure was tested by performing Raman imaging of an isolated nanostructure (end to end coupled nanowire) which is precoated by Rhodamine 6G molecules. This is done with the help of confocal Raman Imaging Microscope (LabRam HR, Horiba Jobin Yvon, France).

For the Raman imaging the AgNW was illuminated with the laser light, wavelength 632.81nm, with polarization parallel to the junction through an objective lens (100x, 0.9NA) in backscattering geometry. For the raman maping (imaging) we choosed the 1361cm^{-1} mode of R6G, which is shown in the figure4 by arrow, with the 5sec/pixel of aquisition time. In this experiment we observed that there is an intense raman signal coming out from the junction part of this nanostructure as compared to the other part which is shown in fig.4(b).

3.2 Studies of light propagation and emission using polarization-controlled illumination

Next for further studies, we tried to see what happen to propagation and emission at the junction and at the distal end when we illumate as a function of polarization. We

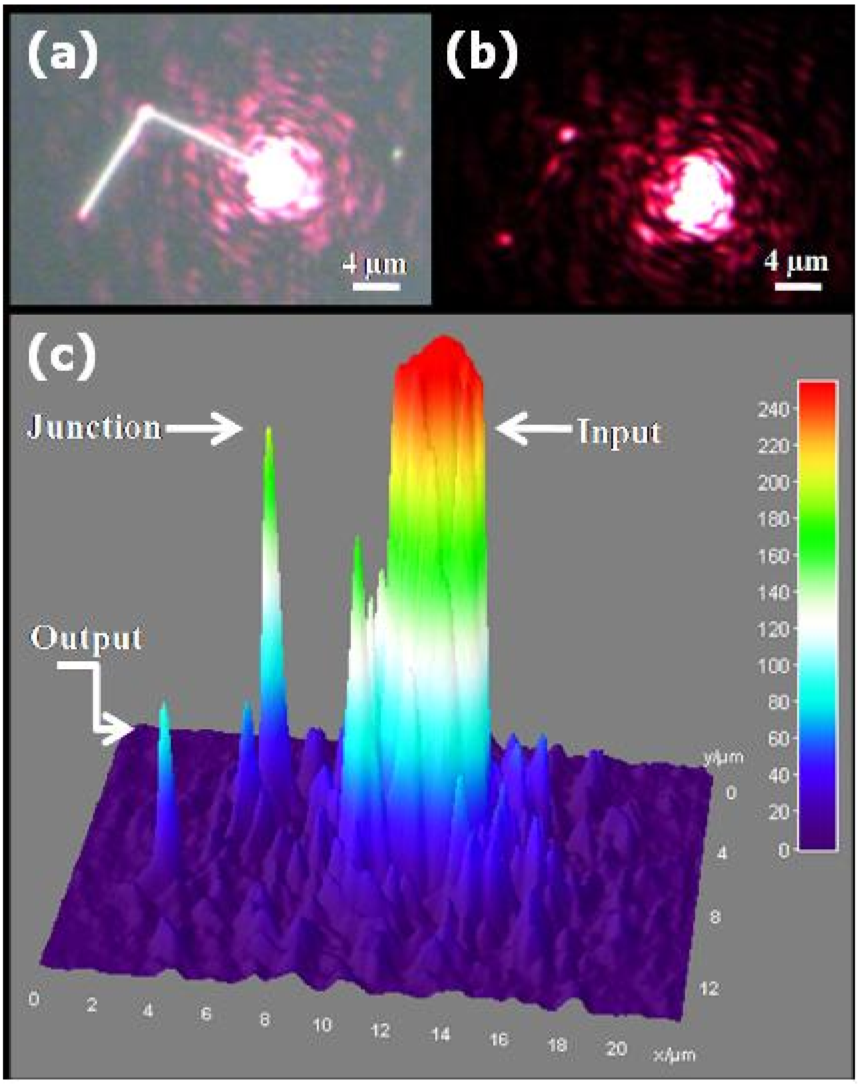

Figure 3: Light propagation and localization in end to end coupled nanowires. (a), (b) shows light propagation and localization in nanowires with and without brightfield illumination, (c) represents 3 – D projection of optical image.[29]

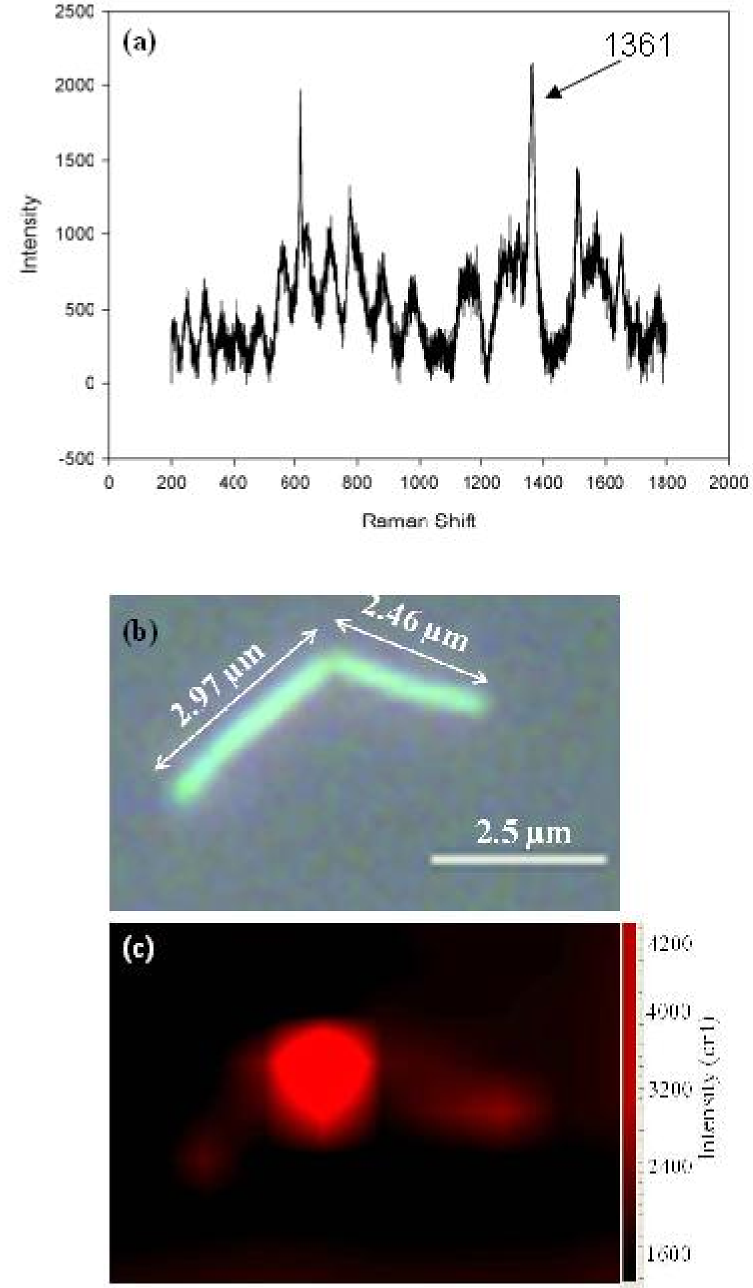

Figure 4: (a) SERS of Rh6G molecule which was coated on AgNWs, (b)An optical image which is used for Raman imaging, (c) Raman image of Ag NW shown in fig.(b). [29]

did two experiment : First, we illuminated at one end of AgNW and tried to find out the emission intensity profile at the junction and at the distal end as a function of polarization, second, we illuminated at the junction and observed what happened to the emission intensity at both the ends of this nanostructure.

3.2.1 Poalrization-controlled illumination at one end of AgNW

We illuminated tightly focused laser light of wavelength 632.81nm through high NA objective lens (100x, 0.9NA) at one end of the AgNW as a function of polarization. We observed that as we change the incident polarization of illuminated light, the emission intensity of junction and distal end is also changed. Fig. 4(e) shows the polar plot of emission intensity of both the junction and distal end as a function of incidence polarization. From the polar plot it can be deduced that the emitted light is maximum when the polarization of illuminated laser is parallel to the illuminated wire axis and emitted light is minimum when polarization of illuminated laser is perpendicular to the illuminated wire axis. Fig.4(a) shows an optical image of AgNW with arrow indicates the intial direction of polarization. The rotation of incident polarization was in anticlockwise direction by taking reference direction in initial direction. Fig.4(b) shows the TEM image of the junction that clearly shows the discontinuity where two wire are joined to each other and fig. 4(c), 4(d) indicate the light propagation through this wires. Fig.4(e) is the polar plot of emission intensity at junction (blue curve) and at distal end (red curve) as a fuction of polarization.

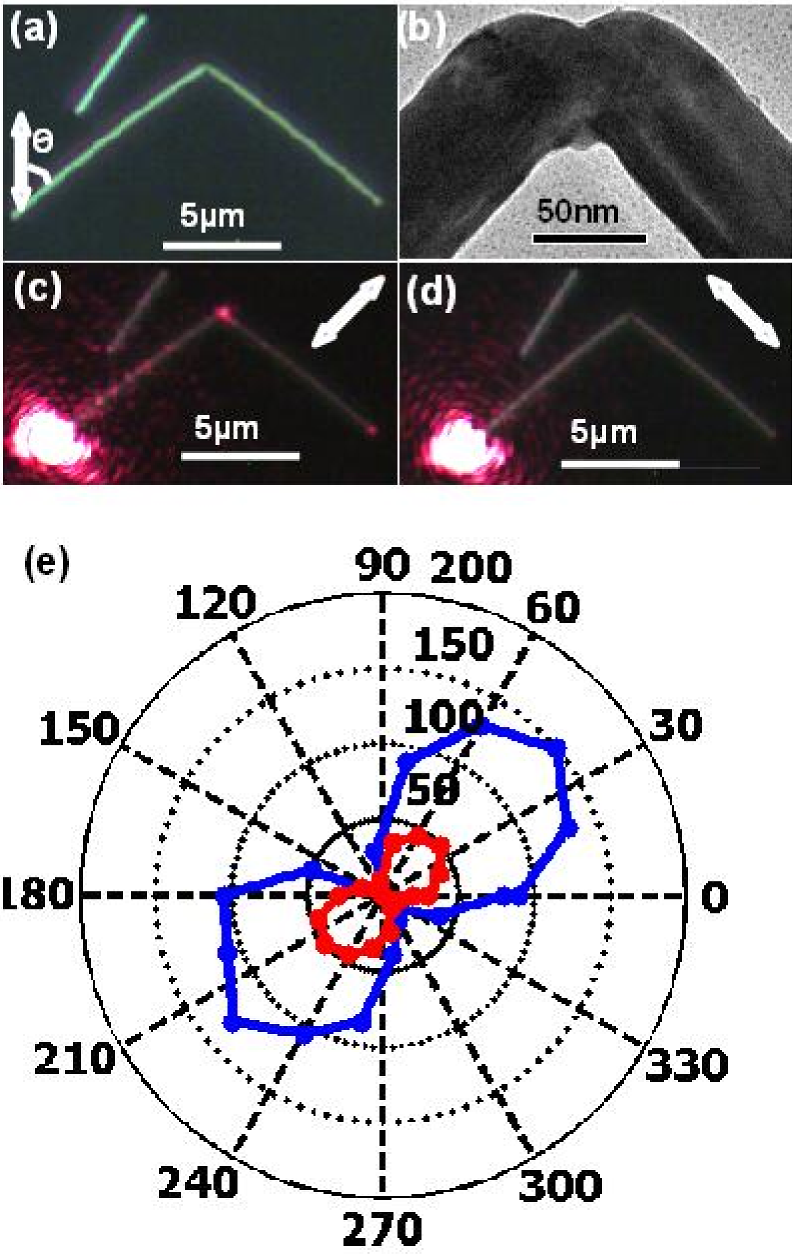

Figure 5: (a) Optical image of end to end coupled nanowire which we have used for polarization controlled illumination with an arrow shows the initial polarization of incident light, (b) TEM image of junction part of nanowire, (c), (d) are laser illuminated optical images with and without brightfield illumination , (e) the polar plot of emission intensity of the junction and the distal end. Image source: [29]

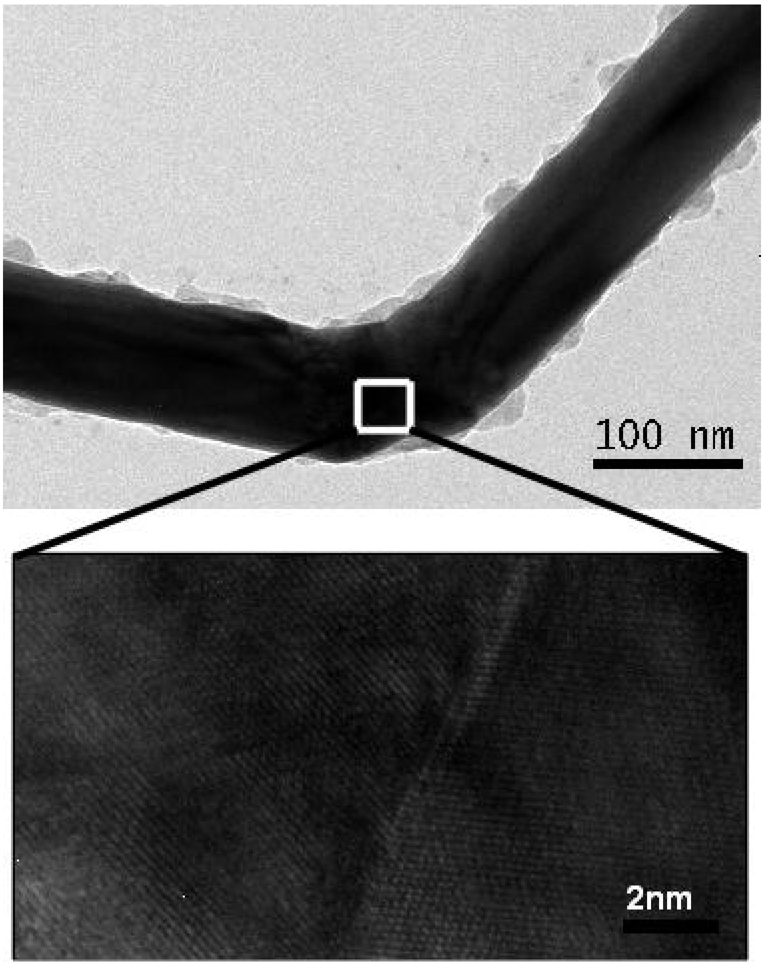

Figure 6: TEM image of end to end coupled nanowire, lower image is HRTEM image of junction at 2nm scale.

3.2.2 Polarization-controlled illumination at the junction

The nanowire was illuminated at the junction as a function of polarization under similar condition described previously, we observed emission from both ends of this nanostructure. Figure6 shows the HRTEM image, at a very high resolution (2nm), of nanowire junction. This attachment between nanowires creates a discontinuity in this nanostructure which caused coupling and decoupling of light.

Fig.7 shows the optical images in the left column and the polar plot of emission intensity of both ends as a function of incident polarization of light in the right column.

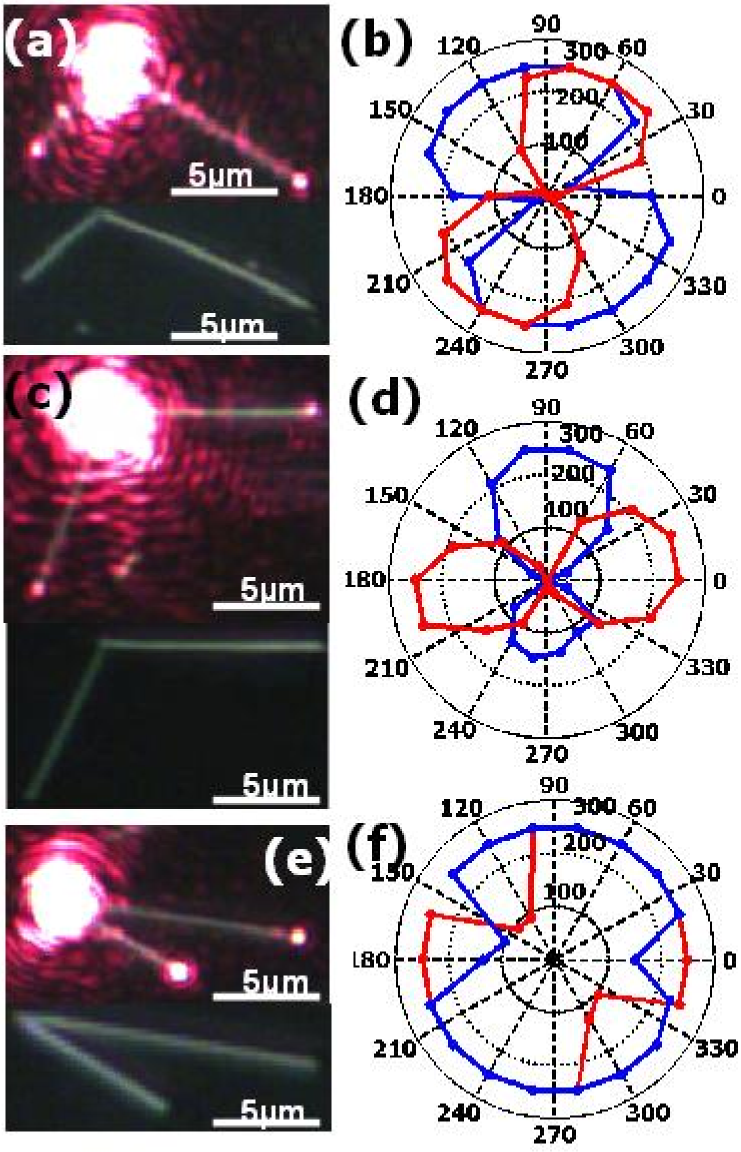

Figure 7: Left column (a),(c),(e) represents the optical image of focused laser beam on the junction in the presence of brightfield illumination, right column (b),(d),(f) is the polar plot of emission intensity of left end(Blue curve) and right end (Red curve).

4 Discussion

For this purpose we have used silver (Ag) nanowire and observed how the light propagated and localized. A special geometry of nanowires was used : end to end coupled nanowires, where two nanowires coupled to each other serially. Using this geometry of Ag NW we did two experiments : First is the studies of light propagation and localization and second is to control the propagation of light in this geometry by controlling the incident polarization of illumination.

4.1 Light propagation and localization in end to end coupled nanowire

End-to-end coupled nanowire were synthesized chemically where two nanowire get attached to each other during chemical reaction. In this way the two single crystal nanostructures attached together and formed a single nanostructure which can be used for nanophotonic applications. We used this nanostructure for testing light propagation and localization. In this way, this unique geometry of nanostructure provides two output sources of photons one at the junction and other at the distal end. This configuration can provide dual advantages of light propagation and light localization in a single nano element. We can use this aspect in nano photonic devices for multiplexers on micro-photonic chips to transfer information simultaneously in two different direction.

4.1.1 Testing of light propagation

For testing the light propagation property of this nanostructure we illuminated tightly focused laser of wavelength 632.81nm through an objective lens (100x, 0.9 NA) at the one end of this nanostructure. We observed that the nanostructure emitted light from the junction and distal ends. The emission from the junction is due the discontinuity shown in fig.5. At the illuminated end light is transformed into surface plasmon polaritons (SPPs) which propagated along the metal – dielectric interface. This propagated SPPs partially get decoupled at discontinuity and is released as free photons and rest of the SPPs propagate along the second nanowire a further decouple as photons at the distal end.

4.1.2 Testing the localization of plasmons at the junction

The localization property is tested by performing Raman imaging of an isolated nanostructure by using surface enhanced raman spectroscopy (SERS) technique [23, 24] . For this we used Rhodamin 6G (R6G) molecules which is precoated on isolated silver nanowire. Experimental setup, for testing localization capability of nanostructure, included a confocal raman imaging microscope (LabRam HR, Horiba Jobin Yvon, France) with high numerical aperture objective lens (100x, 0.9NA) using 632.81nm wavelength laser light. For raman imaging of nanowire we used 1361 cm^{-1} mode of R6G molecule. The polarization of incident light was parallel to the junction of the nanowire. Raman mapping shows intense raman signal from the junction as compared to other parts of the nanowire. This intense signal at the juction indicated that there is an electromagnetic hot-spot which enhances Raman signals of r6G molecules. Literature shows that the juction between nanostructure represents electromagnetic hot spot [25] and Raman signals of molecules can be enhanced at the junction [26].

4.2 Studies light propagation and emission using polarization-controlled illumination

4.2.1 Polarization-controlled illumination at one end of the wire

In this experiments we studied the light propagation in end to end coupled nanowire by polarization controlled illumination at the junction and the distal end. The experimental setup included a microscope with darkfield and bright field illumination, an objective lens (100x, 0.9 NA) , He-Ne laser light of wavelength 632.81nm. A tightly focused laser illuminated one end of nanowire as shown in fig.4(c),(d). We observed the emission intensity of junction and distal end at different orientations of polarization with respect to wire axis. We plotted the emission intensity of the junction (blue curve) and the distal end (red curve) as a function of polarization which shown in fig. 4(e). The observed light emission at the junction and the distal ends with the intensity being greater at the junction. This is due to propagation loss of SPPs. This loss can be due to scattering loss, radiation loss.

4.2.2 Polarization-controlled illumination at the junction

Second experiment consists the illumination at the junction which leads SPPs along both the wires simultaneously. We obtained light emission at both the ends as shown in fig.6(a),(c),(e). Fig. 6 (b),(d),(f) show the polar plot of the emission intensity of both the ends as a function of incident polarization for three different end to end coupled nanowire. The intensity at both the ends varies as we changed the orientation of incident polarization. When polarization or its component was along nanowire of end to end coupled nanowire then maximum SPPs propagate leading to intense emission. Light can be propagate to any one of the wires or through both of them simultaneously just by changing the incident polarization. Thus we are using same geometry, as we used in previous experiment, and propagating light signals to both the wires with different intensities on single input incidence. By this way we can switch light from one wire to other of same nanowire just by changing the incident polarization. We can use this kind of property in micro photonic chips as a demultiplexer which takes single input provides multiple outputs, integrated optical circuits (IOCs) using photon-plasmon waveguide [27, 28] etc.

Conclusion

In this project we have done the plasmon assisted light propagation and localization in end to end coupled nanowire and polarization controlled illumination at junction and at the distal end respectively. Experimentally we have found that the end to end coupled nanowire has light propagation over a distance of 10 to 15µm and localization capibilities. End-to-end coupled Ag NW pairs also facilitate localized SERS hot-spots at their junctions. Such unique geometries are useful in multiplexed nanoplasmonic devices where optical signal transportation and localization can be harnessed on a single chip. The propagation and localization properties of AgNW can be controlled by controlling the local chemical properties. The junction shows SERS hot-spot which indicates that there is enhanced electromagnetic field. In addition to this we analyzed the emission from the ends and the junction of end to end coupled nanowire using polarization-controlled illumination at the juction and the ends. The emission from the junction and ends can be varied by changing the polarization of incident light.

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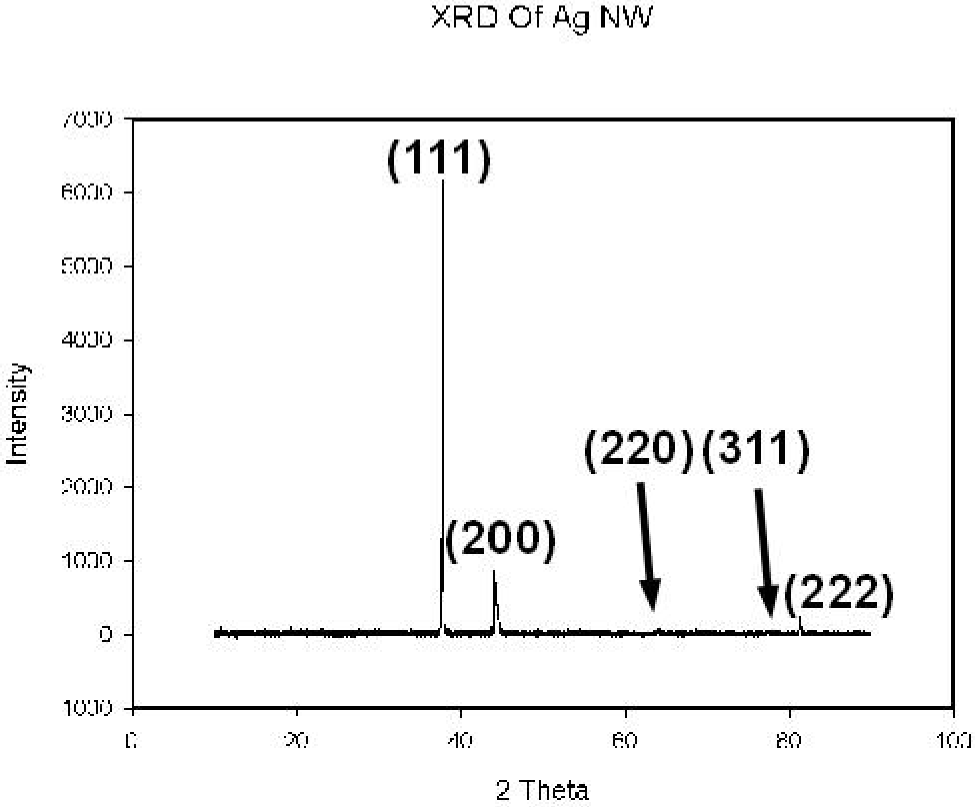
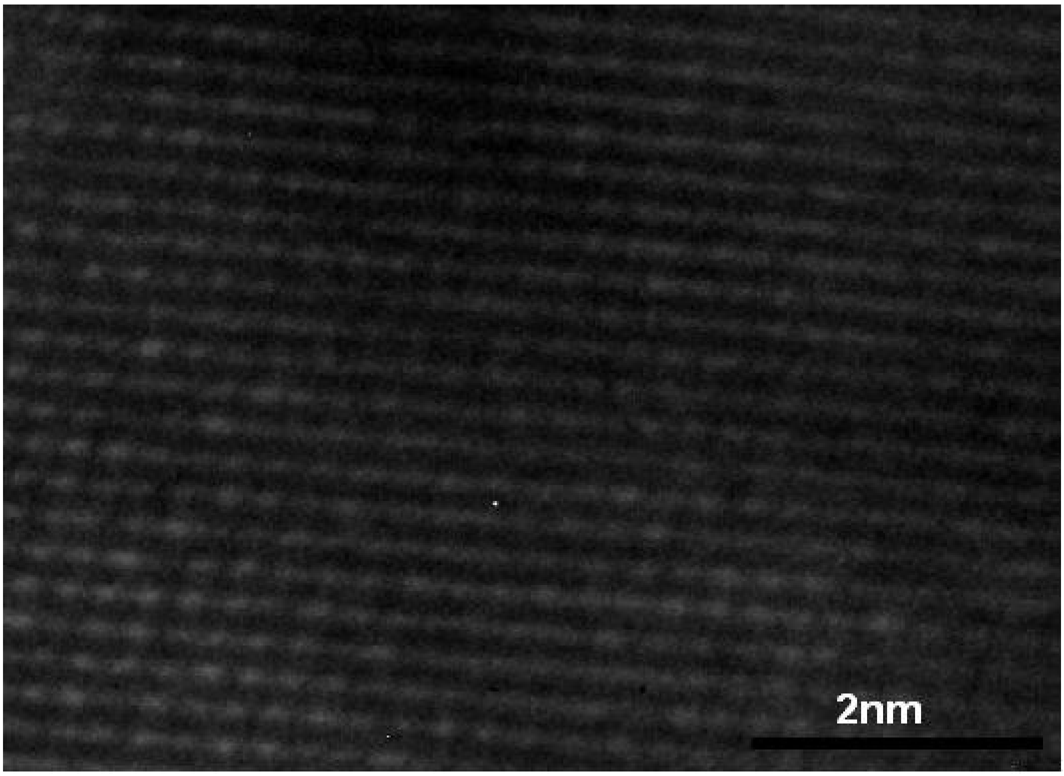
Appendix-A

Characterization of end to end coupled nanostructures

We analyzed end to end coupled nanowires by powder x-ray diffraction (XRD) as shown in fig.2(a), and high resolution transmission electron microscope (HRTEM) in fig.2(c). XRD shows five peaks which indicates that there are five different planes in end to end coupled nanowire. The wires were made up in pentagon shape. These wires are highly crystallined and have FCC lattice structure. The white lines with white fuzzy dots are the crystal lattice planes of nanowires which are visible in HRTEM images.

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