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Photoemission from localized surface plasmons in fractal metal nanostructures

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We use photoemission microscopy to characterize localized surface plasmon distributions in nanostructured gold layers on indium-tin-oxide/glass substrates. The Au films have a fractal dimension of ~ 1.3 and smallest feature sizes of ~ 100 nm. We use femtosecond laser pulses at a wavelength of ~ 800 nm for the plasmon excitation. Photoelectron emission occurs by a three-photon process in localized areas of indium-tin-oxide with ~ 70 nm diameter. In these areas the photoemission rate is enhanced several thousand fold compared to nonstructured surface areas. The results show that plasmon enhanced photoemission can be induced in a nonabsorbing material in proximity to a plasmon-active metal nanostructure. © 2010 American Institute of Physics. [doi:10.1063/1.3457921]

Surface plasmons polaritons have attracted increased research interest due to their potential in concentrating and channeling light at the nanoscale.^{1,2} While plasmon excitations are typically propagating waves, they can be localized in metal films with nanostructured surface, in random geometries, and in islandic metal films.^{3,4} The localization occurs essentially by the confinement of local modes within the nanostructure. This type of localization can lead to a strong enhancement in the local plasmon intensity. “Hot spots” with optical intensities several orders of magnitude higher than the average background intensity have been observed,^{5,6} and are thought to be the source for surface-enhanced Raman scattering, enhanced luminescence, lasing, and other phenomena. Here we report on the possibility to utilize localized surface plasmons for enhanced nanoscale photoemission.

The most effective way to optically excite surface plasmons is by matching the wave vectors of the exciting light and the plasmon modes. In the localization regime the plasmon/light coupling is typically less efficient.³ We show in this paper that photoemission enhancement^{7,8} by several orders of magnitude can occur even under these conditions. We also show that plasmon-enhanced photoemission can occur outside the metallic material supporting the plasmon. We

investigate a configuration where localized surface plasmons in gold lead to photoemission in indium-tin-oxide (ITO) which is transparent to the exciting light and lies in the proximity just outside the gold areas in which the light absorption occurs. Since we are able to resolve the size of the photoemission area, we can provide simple estimates for plasmon transport lengths and plasmon excitation efficiencies in these nanostructures.

200 nm thick smooth Au films were prepared by vacuum evaporation of gold on to 80 nm thick conductive ITO-films on alkali-free glass slides of 0.2 mm thickness. After deposition, the Au films were Ga-milled in a FEI DB237 dual-beam focused ion beam system. Beam currents of 100 pA and beam voltages of 30 kV, were used to mill areas of $10 \times 10 \mu\text{m}^2$ in size to prepare random nanostructures with various degrees of connectivity and feature size. Figure 1 shows results for this process obtained with exposure times of 180, 170, 160, and 150 s. The structure with the strongest photoemission is shown in Fig. 1(c) and has a gold coverage of 50%, a Minkowski–Bouligand fractal dimension of 1.30, a gold border length of $600 \mu\text{m}$, and a smallest feature size of ~ 100 nm. Typically 50 localized emission areas can be identified. Photoemission was also obtained in the other

FIG. 1. Scanning electron micrographs of randomly structured Au films on ITO with Minkowski–Bouligand fractal dimension and Au coverage of (a) 1.36 and 21%, (b) 1.35 and 33%, (c) 1.30 and 50%, and (d) 1.22 and 65%, respectively. The sample in part (c) shows the strongest photoemission.

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FIG. 2. (Color online) (a) Intensity dependence of the photoemission yield for 800 nm light. Inset: illumination scheme for nanostructured Au films on ITO. (b) Photon-energy dependence of the photoemission yield showing a threshold at 3×1.37 eV.

structures shown in Fig. 1 but the emission rates and the number of emission areas are lower.

An aberration-corrected photoemission electron microscope (PEEM) (Ref. 9) was used to visualize the plasmon distribution. The microscope provides total-yield photoemission images employing a frequency-doubled Ar^+ -ion laser or an Hg lamp as ultraviolet light sources. With these sources the photoemission occurs in single-photon processes with linear intensity dependence. Plasmon excitation in the Au was achieved with a tunable Ti-sapphire laser providing 100 fs pulses at 80 MHz at an angle of 60° from the sample surface normal. Back and front side illumination of the Au layer occurs as indicated in Fig. 2(a). Simultaneous illumination by ultraviolet and infrared light allows a precise determination of the hot spot locations.

The tunable pulsed laser provides photon energies between 1.37 and 1.6 eV. Since the work functions for ITO and Au are significantly larger than these energies, multiphoton processes are invoked to explain the observed photoemission. The indicator for multiphoton generation is a superlinear intensity dependence in the photoemission yield. Figure 2(b) shows the emission rate to be proportional to the third power of the laser intensity, indicating that a three-photon process is operative providing energies between 4.1 and 4.8 eV. As shown in Fig. 2(b) the photoemission has a threshold at 4.1 eV which is consistent with the work function of ITO.¹⁰ Typical work function values for Au are 4.5 to 5.2 eV depending on surface conditions.¹¹ Thus photoemission from Au can only occur at the highest energies of the available range or in four-photon processes.

Figure 3 shows a close-up of the photoemission environment. In this particular example a bridge-type geometry is depicted where the planar ITO film connects two separate Au areas with a gap of ~ 100 nm. The composite micrograph clearly shows that the photoemission originates from the ITO film. Since the ITO itself is not absorbing at the primary photon energies, the photoemission must be induced by energy transfer from the absorbing Au film. The diameter of the emission area is typically 70 ± 25 nm as shown in part (b). Comparing the count rates in the bright spots to count rates for unstructured planar areas of ITO films we find photoemission enhancement factors of ~ 6000 for the hot spots. Accounting for the cube-law in the three-photon process, the

FIG. 3. (Color online) (a) Top: Scanning electron microscope (SEM) image of typical hot spot environment, bright areas are Au covered; bottom: composite micrograph of SEM and multiphoton PEEM images of localized photoemission at $\lambda = 792$ nm from ITO bridge between two Au islands. (b) Photoemission count rate across a localized photoemission area. Inset: PEEM image of localized emission. (c) Excitation spectra of various hot spots.

optical intensity enhancement comes out to be ~ 18 .

The spectral properties of the localized plasmon resonances are shown in Fig. 3(c). From the excitation spectrum we find a typical bandwidth of ~ 50 nm, which gives quality factors of 16 to 18. Thus both the q -factor and the intensity enhancement factor are of the same magnitude and consistent with the idea that the emissive areas are localized resonances of the plasmon distribution. The observed intensity enhancement of 18 in an area of 70 nm diameter could be achieved by energy collection over an area with a radius of approximately 150 nm. Such a transport distance is consistent with the localized nature of the plasmon distribution as it corresponds to the smallest feature size in the Au layer structure. A transport distance of 150 nm also lies, as expected, at the very low end of transport ranges of non-localized surface plasmons in Au.¹²

Taking account of the instrument sensitivity we find that the average hot spot delivers only a few hundred electrons per second. The low emission rate can be attributed to the combination of inefficient light-to-plasmon coupling in the random geometry, low probabilities for the three-photon processes, and the short-range plasmon energy transfer. Clearly, all of these factors could be improved significantly. Thus, instead of the random geometry an efficient wave vector matching configuration could be utilized to improve the light coupling. With lower-work-function emitters and lower excitation wavelengths, two-photon processes, rather than three-photon processes could be utilized, and finally, by implementing improved plasmon transfer structures, larger enhancement factors could be achieved. With these improvements considerably more powerful electron sources should become feasible in planar geometry. At the present stage the main attractiveness lies in the fact that simple random structures experimentally permit nanoscale photoemission from low-energy photons. These structures may be useful in signal routing and processing, nanoelectrochemistry, and in nanometrology. For some of these applications it is of impor-

tance that the electron emission can be induced in the near-field proximity of metals, as we showed here.

To summarize, we have shown that locally enhanced photoemission can be obtained in random metal nanostructures. We estimate that the optical intensity in these hot spots is at least a factor ~ 18 higher than the intensity in unstructured films. This is consistent with plasmonic energy transfer distances of ~ 150 nm. The plasmon intensity enhancement translates into a photoemission enhancement of 6×10^3 due to the strong nonlinearity of a three-photon process. In the present study, photoemission is only observed outside the plasmon generating Au film in areas which are covered with ITO. The time-averaged photoemission rates are of the order of a hundred electrons per second. The experimental conditions are of interest for a number of useful applications of this technique in analysis and microscopy.

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