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Exploring the intellectual structure of nanoscience and nanotechnology

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Abstract

Understanding the research trends and intellectual structure of nanoscience and nanotechnology (nano) is important for governments as well as researchers. This paper investigates the intellectual structure of nano field and explores its interdisciplinary characteristics through journal citation networks. The nano journal network, where 41 journals are nodes and citation among the journals are links, is constructed and analyzed using centrality measures and brokerage analysis. The journals that have high centrality scores are identified as important journals in terms of knowledge flow. Moreover, an intermediary role of each journal in exchanging knowledge between nano subareas is identified by brokerage analysis. Further, the nano subarea network is constructed and investigated from the macro view of nano field. This paper can provide the micro and macro views of intellectual structure of nano field and therefore help researchers who seek appropriate journals to acquire knowledge and governments who develop R&D strategies for nano.

Keywords
Nanoscience and nanotechnology

Notes

Noble metal nanoparticles have found wide applications in bioanalytic1 as well as photonic applications owing to their unique light scattering and absorption properties.2,3 These properties arise from resonant oscillations of the conducting electrons of the nanoparticles called localized surface plasmon resonances (LSPRs). The energies of LSPRs lie typically in the optical regime and depend on the nanoparticle size, shape, and composition as well as on the orientation of the electric field relative to the particle and the dielectric properties of the surrounding medium.4,5 The latter dependence of the LSPRs on the nanoparticle surroundings has been exploited to realize a number of labelfree biosensors.614 Persistent efforts have been made to finely tune the composition, shape, and size of metallic nanoparticles and thereby control their optical properties. Recently, a new kind of metallic nanoparticle, called nanostars has been synthesized.15 The nanostars are composed of a central core from which a number of protruding tips extend. They typically show a LSPR of the core and multiple LSPRs corresponding to the tips and coretip interactions. The latter are polarization dependent and accompanied by large local electric field enhancements at the sharp ends of the tips.16 Those locally enhanced fields have been exploited to amplify Raman signals (surface-enhanced Raman spectroscopy, SERS) allowing molecular detection at zeptomolar levels17 and, very recently, have enabled the demonstration of SERS at the single gold nanostar level.18 As a consequence of the confined electric field enhancement at the tips, the spectral positions of the LSPRs of a nanostar are expected to depend strongly on the dielectric environment around the tips. In this paper, we characterize this dependency at the single nanoparticle level and demonstrate its application for the detection of biomolecular interactions using the wellknown streptavidinbiotin assay. RESULTS AND DISCUSSION Single gold nanostars were located and investigated spectroscopically in a darkfield microscope equipped with a spectrometer coupled to a liquid-nitrogen cooled, back illuminated CCD camera. The nanostars present multiple LSPRs. A representative polarization dependent scattering spectrum of a single nanostar is shown in Figure 1C. Multipeak (Lorentzian) fits to these spectra reveal the presence of four resonances. Although the nanostars may present more observable resonances, for consistency we focused our single-particle investigations on nanostars whose spectra could be satisfactorily fitted with four resonances exclusively. Figure 1D shows a scatter plot of the amplitude vs position obtained from 4-Lorenzain fits to the polarization dependent spectra of Figure 1C and is representative of all nanostars investigated. In all cases we observe the nanostars present one weak and nearly polarization-independent resonance *Address correspondence to fernando.stefani@df.uba.ar. Received for review April 12, 2010 and accepted October 06, 2010. Published online October 13, 2010. 10.1021/nn100760f © 2010 American Chemical Society ABSTRACT Gold nanostars provide high sensitivity for single nanoparticle label-free biosensing. The nanostars present multiple plasmon resonances of which the lower energy ones, corresponding to the nanostar tips and coretip interactions, are the most sensitive to environmental changes.

Streptavidin molecules are detected upon binding to individual, biotin-modified gold nanostars by spectral shifts in the plasmon resonances. Concentrations as low as 0.1 nM produce a shift of the tip related plasmon resonances of about 2.3 nm (5.3 meV). at around 540560 nm which we ascribe to the nanostar core. The longer wavelength resonances are polarization dependent and are attributed to the tips or the interaction between tips and core.16 We note the peak at around 610 nm presents a weaker polarization dependency than expected for a tip or coretip interaction resonance. A possible explanation is the spectral superposition of tip or tipcore resonances corresponding to similar, but differently oriented tips. To test the magnitude of the spectral shift of the LSPRs upon bulk changes in the refractive index, we measured the spectra of individual nanostars in air, water (Milli Q), and glucose solutions of different concentrations (Figure 1D). We quantify the shift of each peak with a four-peak (Lorentzian) fit to each spectrum. Since the spectral position of the resonances varies from one nanostar to another, in order to characterize the typical behavior of the

nanostars, we computed the average response of resonances from different nanostars peaking between 650 and 750 nm. The determined average sensitivity is 218 nm/RIU (Figure 1D). Considering the average fwhm of 43 nm for the resonances computed, we obtain a figure of merit (FOM) for the sensitivity of 5, which is within the range of FOM values reported for Au nanostars,15 above Au bipyramids19 and much higher than other Au nanomorphologies (0.6 for spheres, 1.5 for nanocubes, 2.6 for nanorods).19 Figure 2 shows a schematic of the assay. The biotinylated gold nanostars provide a number of binding sites for streptavidin (SA). Figure 3A shows the scattering spectra of a single biotin-functionalized gold nanostar before and after incubation with a 1 m streptavidin (SA) solution (in 0.1 M PBS buffer solution; pH 6.8). When SA binds to the biotin moieties on the nanostar surface, the plasmon resonances shift to lower energies (longer wavelengths) due to the increase in local refractive index.8,14,20 Again in this case we quantified the plasmon shifts with a four-peak (Lorentzian) fit to the spectra of the single nanostars (Figure 3A). The peak labeled as "0" attributed to the plasmon of the nanostar core, is polarization independent and shows the smallest response to SA. We focused on the more sensitive longer wavelength resonances. In this case we quantify the shift in wavelength units (Figure 3A) and observe an unambiguous trend. The longer wavelength (lower energy) peaks respond to the presence of SA target molecules with a stronger shift (Figure 3B) in agreement with the previous reports and theoretical calculations.14,2123 A larger detectable wavelength shift is of valuable practical advantage for biosensing purposes. In addition, we observe the same trend for the relative spectral shift:

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E/E 1.10%, 1.11%, and 1.36% for peaks 1, 2, and 3, respectively, which indicates the lower energy resonances are fundamentally more sensitive to dielectric changes of the environment.24 The specificity of the biotinylated-nanostars was tested in a series of control experiments where again we considered the lowest energy resonance of Figure 1. Structure and scattering of gold nanostars. (A and B) transmission electron microscopy (TEM) images of two gold nanostars. (C) Polarized light scattering spectra of a single gold nanostar immobilized on a glass substrate (in 0.1 M PBS buffer solution; pH 6.8) as a function of the polarization angle (0°180°) of the incident light. The spectra are vertically offset for clarity. A four-peak (Lorentzian) fit to the first spectrum is shown. (D) Scatter plot of the amplitude vs position obtained from 4-Lorenzian fits to the spectra shown in panel C. (E) Average spectral shift of nanostars resonances peaking between 650 and 750 nm as a function of the surrounding refractive index. Data points for air (n 1.00), Millipore water (n 1.33), and 0.35 and 0.95 M glucose water solutions (n 1.35 and n 1.38, respectively). Figure 2. Schematic representation of streptavidinbiotin interactions on a single gold nanostar. ARTICLE www.acsnano.org VOL. 4 ▪ NO. 11 ▪ 6318–6322 ▪ 2010 6319 nanostars lying in the 650750 nm range. Typical control curves are shown in Figure 3B. First, we controlled the stability of the LSPRs in the buffer solution. No shifts are observed (ctrl 1). Second, we tested the nanostars response to 1 m BSA solution. BSA has a molecular weight similar to SA and thus the SA and BSA solutions have comparable refractive indices. Only a very small shift is observed at long incubation times due to unspecific binding (ctrl 2).

Finally, we performed an additional control measurement with gold nanostars modified with nonbiotinylated BSA. In this case, there is an LSPR shift of around 1.5 nm due to the nonspecific adsorption of streptavidin (ctrl 3). We analyzed in detail the response of the different plasmon resonances as a function of the polarization angle of the incident light. The scattering spectra of individual nanostars were acquired for different polarization angles (as in the example shown in Figure 1) before and after the addition of SA. Figure 4A shows the shift of the plasmon resonances labeled as "0", "1", "2", and "3" in Figure 3A as a function of the polarization angle, before and after the addition of 1 m SA. The shift produced upon addition of SA is fairly independent of polarization. In Figure 4B, the plasmon shifts produced on 21 resonances of six different nanostars after incubation with 1 m SA are shown as a function of the spectral position of the resonances.

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10. Baciu, C. L.; Becker, J.; Janshoff, A.; So¨ nnichsen, C. Protein Membrane Interaction Probed by Single Plasmonic Nanoparticles. Nano Lett. 2008, 8, 1724–1728. Figure 5. (A) Real time measurements of the plasmon peak shift of the LSPR “3” of single biotin-modified gold nanostar for different streptavidin concentrations. (B) Average shifts as a function of the SA concentration (five different nanostars for each concentration). The error bars indicate plusminus a standard deviation. The line is a guide to the eye.