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ENHANCING AND LOCALIZING LIGHT-MATTER INTERACTIONS USING SURFACE PLASMONS

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(Invited paper)

ABSTRACT. The enhancement of light-matter interactions using the strong fields associated with surface plasmons in metallic structures is a powerful concept used for example in surface enhanced Raman scattering and plasmonic sensing. We investigated the coupling between nanoscale emitters and surface plasmons in sharp metal tips and smooth metal films by detecting the angular distribution of the emitted radiation. The localized nature of the interaction is utilized in a scanning-probe approach for high-resolution Raman and photoluminescence microscopy of one-dimensional nanostructures.

Localized surface plasmons provide a powerful means to concentrate light far below the diffraction limit and to produce high local field intensities. Compared to conventional optics based on dielectric lenses the excitation probability of nearby quantum systems can thus be enhanced substantially and controlled with nanoscale precision. This phenomena is exploited for example in tip-enhanced near-field optical microscopy [1]. In this approach, a sharp metal probe acts as an optical antenna converting propagation free space radiation efficiently to localized currents and vice versa [2]. In the context of quantum systems, these currents represent excitation and de-excitation. As an example, Figure 1 shows Raman and photoluminescence images of a single DNA-wrapped single-walled carbon nanotube (SWNT) obtained after laser excitation at 632.8 nm and simultaneous detection around 703 nm and 950 nm, respectively. The spatial resolution of the image is limited by the size of the probing metal tip to about 15 nm, far below the limit imposed by diffraction of about 300 nm.

In order to show that coupling to localized surface plasmons in the metal tip indeed results in both enhanced excitation and emission we detected the angular distribution of the radiation with and without tip. Semiconducting single-walled carbon nanotubes were used as one-dimensional model systems. On glass substrates the photoluminescence (PL) from single SWNTs was found to lead to dipolar emission patterns (Fig. 2(a)) as observed before for single dye molecules [4]. Approaching a sharp gold tip antenna to within few



Figure 1. Topography of a DNA-wrapped single-walled carbon nanotube together with the simultaneously acquired near-field Raman and photoluminescence image. The spatial resolution of the optical images is about 15 nm.



Figure 2. Angular photoluminescence radiation pattern detected from a single SWNT on glass without (a) and with metal tip (b). (c) Radiation pattern detected for a single SWNT deposited on a thin gold film. The radiation patterns indicate coupling to localized tip-plasmons in (b) and propagating surface plasmons in (c).

nanometers enhances the PL intensity and results in a substantial redirection of the emission as indicated by the observed radiation pattern (Fig. 2(b), [5]). In the presence of the tip the pattern corresponds to that of a vertical dipole indicating that nanotube PL is emitted efficiently via localized tip plasmons. Based on our experimental data the radiative rate enhancement provided by plasmon coupling and the contribution of excitation rate enhancement can be distinguished [5].

In further experiments the coupling of PL from single SWNTs to propagating surface plasmons in a thin gold film was investigated. Surface plasmons are detected through leakage radiation that is converted into propagating light using an oil immersion objective. Due to the dipolar radiation characteristics of nanotubes, propagating surface plasmons are launched in this case predominantly in the direction of the SWNT axis (Fig. 2(c)).

Our results underline the potential of plasmonic enhancement of light-matter interactions. While localization forms the basis of high-resolution microscopy, coupling to propgating plasmons can be used to reshape the angular distribution of emission.

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