Diamond nanoparticles as surface-plasmon launchers: towards a « deterministic » quantum plasmonics

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Abstract

A nanodiamond hosting two NV centers is attached onto the apex of an optical fiber-tip and illuminated by a laser light guided by the fiber itself. Gold films are dipped into the optical near field of the functionalized tip. The fluorescence light generated by the NVs efficiently launches surface plasmons that are imaged by near-field microscopy. Since the nanodiamond is a quasi single-photon source, we argue that single surface plasmons form the experimental images. This is a first step towards a "deterministic" quantum plasmonics where quantum plasmons can be launched at any freely chosen position in a plasmonic receptacle.

1. Introduction

The miniaturization of electronic and optical devices faces the crucial obstacle of energy transfer and communication between functional objects at the nanoscale. In the last decade, the use of plasmonics skills has been initiated to challenge this issue. Surface-plasmon polaritons (SPPs) [1,2], i.e. hybrid electron-photon modes propagating at the interface between a metal and a dielectric material, are naturally well adapted to low-dimensional operation. Indeed, as surface waves, SPPs are exponentially damped in the perpendicular direction and can be guided over micron distances in structures laterally miniaturized to sub-wavelength dimensions [1,2]. These attractive properties make SPPs promising candidates for nanoscale optical addressing [3] or light manipulation by tailored plasmonic components, such as prisms and lenses [4] or multiplexers [5].

Previous studies have shown that SPPs can be launched in a metal film by the evanescent light emanating from an optical tip for near-field scanning optical microscopy (NSOM) coupled to an excitation source of appropriate wavelength [6-8]. This is because this near field is made of evanescent waves that possess high spatial frequencies able to couple to SPPs. This is in contrast with far-field (propagating) photons, which do not have enough momentum for that. Here we briefly review our recent experimental work aimed at demonstrating that the near-field fluorescence emanating from diamond nanocrystals attached onto an optical tip could also efficiently launch SPPs on a metal film [9]. This should allow for a tighter spatial control on the SPP injection position and opens the way to a "deterministic" plasmonics, or even quantum plasmonics, if a single diamond nanocrystal hosting a single quantum emitter is used to launch SPPs one at once.

2. Attaching a nanodiamond onto an optical tip

Our method for attaching a well-selected nanodiamond onto an optical tip has been described in detail in previous papers [10,11]. Briefly, nanodiamonds with an average size of 25 nm are dispersed on a fused silica cover slip. Part of the nanodiamonds is fluorescent [12] thanks to nitrogen-vacancy (NV) color-centers that the particles can contain following their production process (see. e.g. [13]). The fluorescent nanodiamonds are located by means of fluorescence NSOM with the optical tip used in the so-called excitation mode. They can be characterized by second-order time-intensity correlation measurements [14] to determine how many NVs they contain, generally one or two in the particular diamond sample that we used extensively. Once an "interesting" nanodiamond (small size, bright fluorescence, if possible single NV occupancy) has been located, the NSOM tip is surreptitiously approached during scanning to this particular diamond. This produces trapping of the particle by the optical tip. Trapping occurs

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because the tip has first been coated with a positively charged polymer, whereas the diamond bears a surface negative charge because of its production process. The graft event is confirmed to have taken place because it translates into a persistent increase of the fluorescence signal embarked by the scanning tip.

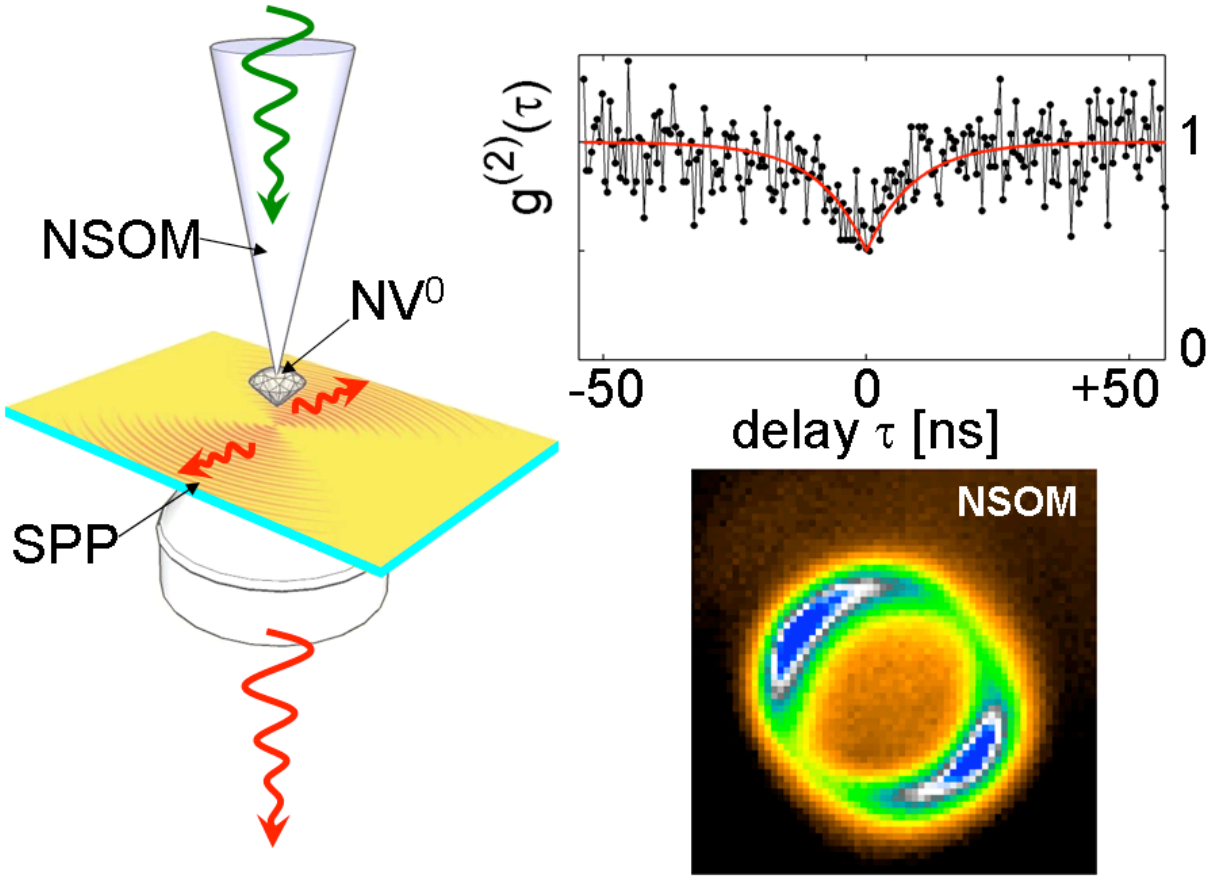

Figure 1: *Principle and demonstration of SPP launching by a nanodiamond-based tip. Left: A single diamond nanocrystal with two NV centers is glued at the apex of a bare fiber tip. The sole fluorescence of the color centers is coupled to SPPs propagating along a gold film. Light transmitted trough the film either by radiation leakage into the substrate or diffraction of the plasmon field by a nanostructure, e.g. a slit in the film, is collected by a microscope objective for NSOM imaging. Right, upper curve: A typical intensity-time second-order correlation function of the two color centers at the tip apex showing the quantum nature of the fluorescence light. Right, bottom image: A typical NSOM image of a gold nanostructure – a 2 micron circular slit in an opaque gold film - acquired with a quantum plasmonic tip. Reprinted from [9].*

3. Launching surface plasmons with a diamond-based tip

The preceding diamond-functionalized tip is used to launch SPPs into the gold film as shown schematically in Fig. 1. For that, the ND is approached to a few tens of nanometers to the gold film and excited with a blue-green light at 488 nm injected into the fiber. The part of its red-orange fluorescence light that is transmitted through the metal film by light leakage in the silica substrate, in the case of a thin (60 nm) film, or by diffraction through a sub-wavelength slit in a thick opaque (200 nm) film is imaged either by leakage radiation microscopy [15] or by NSOM. An example of NSOM imaging of a ring-like aperture slit (1.8 micron inner diameter, 150 nm rim width) is shown in Fig. 1. Here the diamond hosts two NV centers as demonstrated by the correlation curve of Fig. 1, which depicts a distinctive photon-antibunching phenomenon down to 0.5 at zero delay, i.e. $g^{(2)}(0)=1/2$. Clearly, the optical image shows that there is plenty of light inside the cavity although the gold film is opaque. Such a behavior is not observed if the image is recorded at the excitation wavelength of 488 nm [9], which is indeed inefficient to excite SPPs into

gold because of a strong interband absorption in gold in the red part of the spectrum. The interpretation of the NSOM image is that SPPs are excited by the fluorescence light of the ND and propagate toward the rim zone where they are diffracted back to a photon signal that is collected on the other side of the thick opaque film to form a transmission image.

Three features of the experiment are worth commenting on. First, the NSOM image of Fig. 1 is obviously polarized. We showed in [9] that this polarization does not coincide with that of the excitation. This is because the NV quantum emitters have a well-defined orientation within the ND crystal (Note however that the precise orientation of the transition dipoles cannot be controlled within our ND attachment procedure). Second, the SPP launching position coincides with the tip position that is tightly controlled in a NSOM microscope, so that it can be tightly controlled too. This can be very useful in further studies. Third, we point out that since $g^{(2)}(0)=1/2$ the launching source cannot emit more than two photons and consequently launch more than two SPPs at once. This is a particle-like behavior that cannot be understood in the context of classical optics. Hence, the NSOM image of figure 1 is a manifestation of the wave-particle duality for SPPs. This entails that the quantum plasmonics realm has indeed been entered [16].

4. Conclusion

The possibility of launching SPPs in a very limited number, down to only one if a nanodiamond with single NV occupancy is used as launcher, at well chosen positions opens new perspective in the emerging film of quantum plasmonics. For instance, some groundbreaking experiments in quantum optics [17] could possibly be extended to surface plasmonics. Another perspective, among others [9], is the investigation of qubit-qubit entanglement mediated by plasmons [18].

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