Ultrabright Room-Temperature Emission from Single Plasmon-Enhanced Nitrogen-Vacancy Centers in Diamond

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Abstract: We report unprecedentedly bright fluorescence from single nitrogen-vacancy centers in nanodiamonds. The intensity enhancement is achieved by placing nanodiamonds in gaps between a metal film and randomly dispersed metal nanoparticles. The shortening of fluorescence lifetime below 1 ns combined with an efficient outcoupling of plasmons into the far-field lead to a detected fluorescence intensity in the range of 10^7 counts per second (cps). The photostability of nitrogen-vacancy centers allows reaching the saturation regime and estimating the total brightness enhancement resulting from the use of plasmonic nanostructures.
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1. Introduction

The fluorescence intensity from single solid-state emitters can be dramatically enhanced by engineering the electromagnetic local density of states. In dielectric resonators, this enhancement is achieved by high quality factors, while in plasmonic nanostructures it relies on highly subwavelength mode volumes. The latter approach ultimately promises higher emitter brightness [1], with the caveat that high ohmic losses in the metal must be circumvented.

Nano-patch antennas (NPAs) are plasmonic nanostructures formed by a metal film, a dielectric spacer layer and a plasmonic nanoparticle. NPAs simultaneously offer a high local density of states in the gap between the film and the nanoparticle and an efficient outcoupling of gap plasmon energy into the far-field [2]. Recent results [3] show that NPAs can greatly shorten the fluorescence lifetime of single quantum emitters such as colloidal quantum dots. However, in order to understand the full potential of NPAs for enhancing the quantum emitter brightness, it is necessary to employ photostable emitters and explore the fluorescence intensity in the saturation regime.

In this work, we utilize nitrogen-vacancy (NV) color centers in diamond [4] – bright and photostable single-photon emitters, which can be found in diamond nanoparticles with sizes down to a few nm. The NPAs are formed by a silver film, an approximately 6 nm thick polymer spacer layer, and randomly positioned silver nanocubes (see Fig. 1(a)) with a dimension of 100 nm. Nanodiamonds with an average size of 20 nm and containing NVs were randomly dispersed directly atop the spacer layer resulting in some of them becoming trapped between the film and the subsequently dispersed nanocubes.

2. Experimental results

We compare the emission from control NVs in nanodiamonds placed on glass substrate to that of NVs enhanced by NPAs. In both cases, we examined a collection of 14 emitters and data for one representative emitter from each set are plotted on Fig. 1. Due to the highly confined gap plasmonic modes, the fluorescence lifetime of NVs in NPAs is shortened by two orders of magnitude compared to that of NVs on glass (Fig. 1(b)). For the NPA-enhanced NV, we observe a bi-exponential decay with a faster ($\tau_1 \sim 10\,\text{ps}$) and a slower ($\tau_2 \sim 100\,\text{ps}$) component, indicating the presence of two different emission processes. The longer time constant τ_2 is on average over 100 times shorter than the fluorescence lifetime of NVs on glass (0.47 vs 62 ns). Owing to the efficient outcoupling of plasmons into the far field, the detected emission brightness of the NV in NPA reaches several Mcps (Fig. 1(c)). The NVs on glass are analyzed through an oil objective in the total internal reflection (TIRF) mode (NA_{oil} = 1.49), while the NVs in NPAs are analyzed through an air objective (NA_{air} = 0.9). Even though the oil objective offers better collection efficiency than the air objective, the saturation intensity of the NPA-enhanced NVs is over 100 times higher on average than that of the NVs on glass (37 vs 0.26 Mcps). Detected photon rates from NPA-enhanced NVs reach up to 50 Mcps.

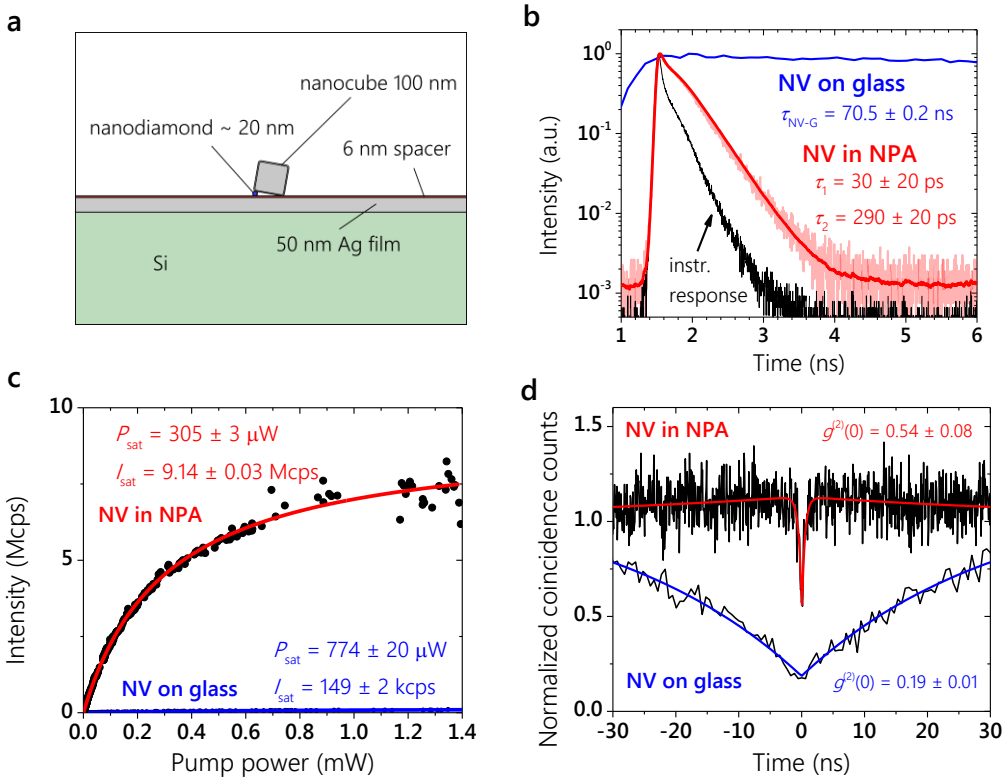

Fig. 1. (a) Schematic representation of a nanodiamond inside a silver NPA. Comparison of (b) fluorescence decays (c) fluorescence intensity saturation curves and (d) autocorrelation curves for representative cases of NV on glass substrate (blue) and in an NPA (red).

Autocorrelation of the intensity signal from the NPA-enhanced NV exhibits a narrow antibunching dip (Fig. 1(d)), confirming sub-nanosecond emission. For the NVs on a glass substrate, the single-photon purity is limited by the background emission from the glass coverslip. For the NPA-enhanced NV, the depth of the autocorrelation dip is limited either by an ultrafast component of NV emission or by the background emission from the NPA. We note that better photon purity at the expense of a smaller increase in brightness can be obtained by using a dimer antenna assembled from silver nanocubes on a dielectric substrate [5].

These results unveil the potential of NPAs for applications in quantum photonics and sensing. In particular, the development of NV-based magnetometry and room-temperature single-photon sources could benefit from the ultrabright fluorescence brought about by such plasmonic nanostructures.

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3. References

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