We report on the controlled coupling of a single nitrogen-vacancy (NV) center to a surface plasmon mode propagating along a chemically grown silver nanowire (NW). We locate and optically characterize a single NV center in a uniform dielectric environment before we controllably position this emitter in the close proximity of the NW. We are thus able to control the coupling of this particular emitter to the NW and directly compare the photon emission properties before and after the coupling. The excitation of single plasmonic modes is witnessed and a total rate enhancement by a factor of up to 4.6 is demonstrated.

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Strong enhancement of the fluorescence and scattered light from a single molecule, quantum dot, or nitrogen-vacancy (NV) center can be observed by placing the particle in the vicinity of a metallic nanosphere [1–5]. This enhancement arises due to the excitation of strongly confined and localized surface plasmon on the metallic sphere. Despite the strong enhancement of emissive processes, the resulting photon emission in those realizations does not couple preferably to a dedicated spatial mode and are thus not directly suitable for applications in, for instance, quantum information processing where the coupling between a single spatial mode and a single emitter is often a requirement for scalability.

To obtain an efficient coupling to a specific spatial mode while exploiting the enhancement properties of strongly confined surface plasmons, it has been suggested to use propagating plasmons on cylindrical wires as opposed to localized plasmons [6]. By using this method, it is possible to enhance the emissive process into one particular propagating plasmon mode which can be further transferred into a photonic mode of an optical waveguide with high efficiency [7,8]. Such a light-matter interface mediated by surface plasmons can be used to efficiently generate single photons and to enable strong nonlinearities at the single photon level which can be exploited to make a single photon transistor [9] or to perform a near-error-free deterministic Bell measurement [10].

The coupling of individual single photon emitters to propagating surface plasmon modes on individual silver nanowires (NWs) has been demonstrated for CdSe quantum dots [11] and for NV centers in nanocrystal diamonds [12]. In these experiments, however, the wire-crystal systems were not deterministically assembled thus rendering the rate enhancement estimation highly uncertain. By comparing an ensemble of uncoupled single emitters with an ensemble of coupled single emitters, they estimated rate enhancement factors of 1.7 [11] and 2.5 [12].

In this Letter, we demonstrate the controlled coupling of a single NV center in a diamond nanocrystal to a surface plasmon mode propagating along a silver NW. The wire-crystal system is deterministically assembled by the use of an atomic force microscope. This approach allows us to directly compare the emission properties of a particular NV center in a homogeneous dielectric environment with the emission properties of the same emitter but placed in the near vicinity of a silver NW supporting a propagating surface plasmon mode. The coupling to the propagating surface plasmon mode is evidenced by a decrease in the emitter lifetime and the emission of a single photon is verified by a measurement of the second-order correlation function.

Our experimental setup is a home-built inverted scanning confocal microscope using an oil immersion objective with a numerical aperture of 1.4 combined with a coaxially aligned atomic force microscope (AFM, NT-MDT SMENA), as illustrated in Fig. 1. For the excitation of NV centers we either use a continuous wave or a pulsed laser, both operating at a vacuum wavelength of 532 nm. The pulsed laser has a repetition rate of 5.05 MHz and a pulse width of 4.6 ps. The fluorescence emission from the sample is collected by two optical channels and detected with avalanche photo diodes APD1 and APD2, respectively. The channel of APD1 is directly aligned with the pump beam, while the channel of APD2 is decoupled from the pump beam via a galvanometric mirror, as illustrated in FIG. 1 (color online). Experimental setup: TAC—time to amplitude converter, APD1 and APD2—avalanche photo diode detection channel 1 and 2, respectively, g—galvanometric mirror, BS—50/50 beam splitter, and HWP—half waveplate. The transmitted spectrum of the fluorescence filter F lies between 647 and 785 nm.
Fig. 2 (color online). AFM topography of the NW and the nanodiamond containing the investigated NV center (white circle). The inset shows the height profile of the wire and the diamond, taken along the white dashed line.

FIG. 3. Fluorescence lifetime measurement of the uncoupled NV-center in a uniform dielectric environment (black dots) and of the same NV center after it has been coupled to the propagating SPP mode of the silver NW (open circles).
couples to the propagating plasmonic mode of the silver NW [6].

Evidence for the excitation of the propagating plasmonic mode is given by the fluorescence image recorded with APD2, which is shown in Fig. 5(b). This image has been obtained by continuously exciting the NV center while scanning the image plane using the galvanometric mirror. Two emission spots can be seen in Fig. 5(b). The emission spot “A” in Fig. 5(b) comprises the radiative emission from the NV center together with emission from the nearby NW end face. Spot “B” in Fig. 5(b) only comprises emission from the far NW end face. The intensity measured from spot B is thus proportional to the coupling of the NV center to the propagating plasmonic mode [6,11,12]. However, an exact estimation of the NV center coupling to the propagating plasmonic mode by measuring the intensity of spot B is difficult due to plasmon propagation losses along the NW and the complex reflections at the NW end face in conjunction with the broad emission spectrum of the NV center.

The $g^{(2)}(\tau)$ function measurement with the detectors APD1 and APD2 being both aligned on emission spot A is shown by the open dots in Fig. 4. With the open triangles we present the $g^{(2)}(\tau)$ function measurement between emission spot A and B. The green and blue line represent a best fit to the data for both measurement realizations. In both cases, a $g^{(2)}(0) < 0.5$ confirms the generation of single photons, and the excitation of single propagating surface plasmon modes is confirmed by the measurement between spot A and B.

Finally, we compare our experimental results with theoretical predictions [6]. In Fig. 6 we plot the total rate enhancement for the relevant range of NW diameter and emitter distance from the NW surface. The calculation is done for a vacuum wavelength of 700 nm and we use the Drude model to estimate the electric permittivity $\epsilon_{Ag}$ of silver [19]. In order to account locally for the relatively high refractive index of diamond and for the glass substrate, the electric permittivity of the dielectric medium $\epsilon_{1}$ surrounding the NW was set to 3 in the calculation. For the NW diameter of 55 nm and a diamond height of 27 nm we expect from these calculations a total rate enhancement of 3.8, assuming that the NV center is located at the maximum position of 27 nm away from the NW surface and that the NV centers dipole moment is aligned parallel to the radial electric field component $E_{r}$ of the propagating plasmonic mode. This expected rate enhancement is in good agreement with our experimental result of $3.6 \pm 0.1$.

FIG. 4 (color online). Second-order correlation function measurement of the uncoupled NV center (black dots) and the coupled NV center with both detectors aligned to spot A (open dots) and APD1 aligned to spot A and APD2 aligned to spot B (open triangles). The lines are a best fit to the data: red—uncoupled, green—coupled AA, and blue—coupled AB.

FIG. 5 (color online). (a) AFM image taken after the nanodiamond has been located near the wire. The location of the diamond is indicated by the white arrow. (b) Photoluminescence image of the coupled NV-center-NW system, taken while continuously exciting the NV center, scanning the sample with the galvanometric mirror and recording the signal with APD2. Encoded in the color scale is the APD signal in counts/s.

FIG. 6 (color online). Total rate enhancement as a function of NW diameter and NV center distance from the NW surface $d'$. The open white circles indicate the minimum expected total rate enhancement for each NW diameter and diamond height, the black filled dots are the experimentally measured rate enhancement factors for each NW diameter, and the white arrows are a guide to the eye to indicate data related via the same NW diameter.
The excitation of a single surface plasmon, confirmed by successfully assembled. For all realizations we observe ters for various NV-center–NW systems that we have 0/C28 mode was witnessed by strong rate enhancement factors the plasmon electric field along the NW axis. We emphasize that only for parallel alignment of the NV moment is aligned parallel to calculated under the assumption that the NV centers dipole expected rate enhancement in column four of Table I, obtained rate enhancements we also present the minimum expected rate enhancement in column four of Table I, calculated under the assumption that the NV centers dipole moment is aligned parallel to Ez. All measured and calculated enhancement factors are graphically illustrated in Fig. 6.

In view of improving the coupling efficiency of the NV center to the plasmonic mode it is necessary to decrease both the diameter of the NW and the size of the diamond, as can be seen from Fig. 6. Thinner NWs can, for instance, be obtained by an optimization of the NW fabrication process [14]. As an alternative, it might also be possible to sculp- ture NWs from chemically prepared metallic flakes using focused ion beam milling [20]. We have found experimen-tally, that NWs made by electron beam lithography and thermal metal deposition are not suitable for coupling single NV centers to their propagating plasmonic mode. Fluorescence from those NWs largely overlaps with the emission spectrum of a NV center, which as a direct consequence limits the possibility of detecting single photons. Surface roughness of lithographically prepared NWs further limits the propagation distance of plasmonic modes [21]. Nanodiamonds with sub-10 nm diameters containing single NV centers with stable photon emission rates have recently been reported in the literature [22, 23].

In conclusion, by the aid of an atomic force microscope we have nanoassembled a system comprising a single NV center in a nanocrystal diamond and a chemically grown silver NW. This method allowed us to directly compared the emission properties of a single NV center in a uniform dielectric environment with the emission properties of the same emitter coupled to the NW. An enhancement of the NV centers decay rate by a factor of 4.6 is directly measured and the excitation of single surface plasmons is evidenced by the observation of single photon emission at the far end of the NW. We believe that the method presented in this article in combination with smaller diamonds and optimized metallic structures will pave the way for strongly coupled plasmonic systems.

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<table>
<thead>
<tr>
<th>NW diameter (nm)</th>
<th>Diamond height (nm)</th>
<th>Total rate enhancement τ₀/τ₁</th>
<th>Min. expected rate enhancement</th>
</tr>
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<tbody>
<tr>
<td>65</td>
<td>45</td>
<td>4.6 ± 0.1</td>
<td>2.2</td>
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<tr>
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<td>2.9 ± 0.1</td>
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<tr>
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<td>5.7</td>
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<tr>
<td>50</td>
<td>35</td>
<td>4.2 ± 0.1</td>
<td>2.8</td>
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