Journal of Luminescence 130 (2010) 1628–1634

Contents lists available at ScienceDirect

Journal of Luminescence

journal homepage: www.elsevier.com/locate/jlumin

Controlled coupling of NV defect centers to plasmonic and photonic nanostructures

Michael Barth*, Stefan Schietinger, Tim Schröder, Thomas Aichele, Oliver Benson

Institute of Physics, Humboldt-Universität zu Berlin, Hausvogteiplatz 5-7, D-10117 Berlin, Germany

ARTICLE INFO

Available online 21 December 2009

Keywords:
Defect centers
Diamond
Plasmonic nanoparticles
Microsphere resonators
Photonic crystal cavities

ABSTRACT

Nitrogen-vacancy (NV) defect centers in diamond have recently emerged as promising candidates for a number of applications in the fields of quantum optics and quantum information, such as single photon generation and spin qubit operations. The performance of these defect centers can strongly be enhanced through coupling to plasmonic and photonic nanostructures, such as metal particles and optical microcavities. Here, we demonstrate the controlled assembly of such hybrid structures via manipulation with scanning near-field probes. In particular, we investigate the plasmonic enhancement of the single photon emission through coupling to gold nanospheres as well as the coupling of diamond nanocrystals to the optical modes of microsphere resonators and photonic crystal cavities. These systems represent prototypes of fundamental nanophotonic/plasmonic elements and provide control on the generation and coherent transfer of photons on the level of a single quantum emitter.

1. Introduction

Due to their unique optical properties, nitrogen-vacancy (NV) defect centers in diamond are appealing candidates for the realization of solid-state devices for optical quantum information processing [1,2]. Their excellent photostability [3] and ultralong spin coherence times [4] have been exploited in a number of experiments demonstrating single photon generation even at room temperature [5,6], coherent population trapping [7], and optical readout and manipulation of single nuclear spins [8–10]. In order to utilize these properties for quantum information applications and to further enhance the performance of NV centers, optical coupling to appropriate plasmonic or photonic structures (such as cavities or waveguides) [11,12] is necessary, e.g., to provide control on the generation and coherent transfer of photons in these systems. Here, we will review recent efforts on achieving such a coupling in a well-controlled fashion.

In general, two different mechanisms can be exploited to manipulate and control the optical properties of NV centers, namely the coupling to surface plasmons [13] (e.g., in metal nanostructures) or the coupling to the modes of optical microcavities or waveguides [14]. Both approaches can, in principle, be used to enhance the photon excitation and/or emission rate, to achieve strong coupling between the zero phonon transition and the photon field, and to facilitate an efficient transfer of photons from/to the NV center. Some of the optical characteristics of single NV centers are summarized in Fig. 1, which show a typical fluorescence spectrum (Fig. 1(a)) and the normalized autocorrelation function $g^{(2)}(\tau)$ measured by a Hanbury-Brown and Twiss (HBT) setup (Fig. 1(b)).

Plasmonic nanostructures can act as optical nanoantennas, leading to highly localized photon fields and therefore to a broadband enhancement of the excitation as well as of the radiative and non-radiative decay rates of nearby emitters [15–18]. An interesting aspect is also that photons can be converted into surface plasmon polaritons, guided along plasmonic waveguide structures, and be converted back into photons [19,20]. In order to establish such a photon–plasmon interface with NV centers, a precise positioning of the NV center with respect to the plasmonic nanostructure is necessary. We tackle this problem by using nanomanipulation techniques [21,22] to gain control on the mutual position and coupling of both constituents, as will be described in detail in Section 2 [23].

Contrary to the broadband field enhancement present in plasmonic structures, optical microcavities provide spectrally narrow resonances with high Q-factors. Combined with small modal volumes, these resonances are well suited to achieve a coherent coupling between the zero phonon transition and the optical mode [11], which is required for the observation of cavity quantum electrodynamic (QED) effects and the realization of interfaces between stationary and flying qubits in quantum information processing devices [2]. In principle, an appropriate microcavity can directly be realized in diamond [24,25], but first experimental demonstrations with microdisk resonators [26] and photonic crystal (PC) cavities [27] suffered from large absorption and scattering losses owing to the poly-crystallinity of the

* Corresponding author. Tel.: +49 30 2093 4941; fax: +49 30 2093 4718.
E-mail address: michael.barth@physik.hu-berlin.de (M. Barth).

0022-2313/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.jlumin.2009.12.004

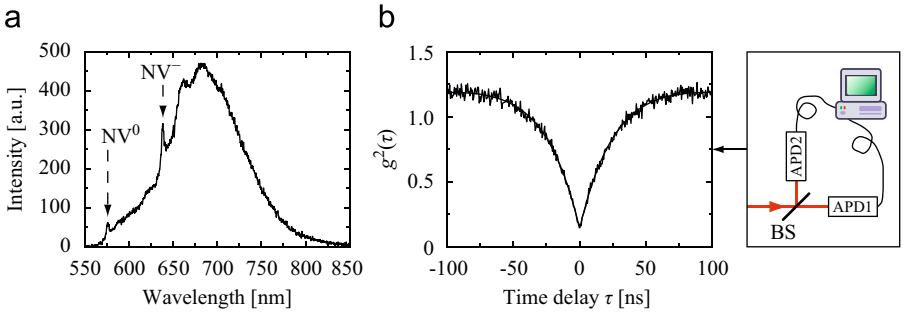

Fig. 1. (a) Typical fluorescence spectrum of a single NV center in a diamond nanocrystal at room temperature. Two zero phonon lines can be observed, associated with the neutral (NV^0) and negatively charged (NV^-) state of the NV center. (b) Second order correlation function $g^{(2)}(\tau)$ (left) of the same NV center, measured using an HBT setup (right). The dip at $\tau=0$ clearly indicates the single photon character of the emission. The HBT setup consists of a beamsplitter (BS) and two avalanche photodiodes (APDs).

employed diamond material. Corresponding implementations with single-crystalline diamond [28] are very challenging and have not yet been realized. An alternative approach is the use of resonators made from non-diamond material to which NV centers are coupled. Such hybrid devices can either incorporate single-crystalline diamond films [29–31] or diamond nanocrystals [32–34]. Using bulk diamond has the advantage of superior optical properties of the NV centers, as nanocrystals can induce inhomogeneous broadening and spectral diffusion of the zero photon transition [35]. However, coupling of a single NV center in such a structure has not yet been demonstrated. In our studies we use diamond nanocrystals containing single NV centers. Again, we exploit nanomanipulation techniques to demonstrate the controlled coupling to microsphere resonators [36,37], as described in Section 3.1, and to PC cavities [38,39], as described in Section 3.2.

2. Coupling of NV centers to plasmonic nanoparticles

In this section we review the experimental realization of a controlled coupling between diamond nanocrystals (20–35 nm in height, purchased from Microdiamant) and spherical gold nanoparticles (60 nm in diameter, purchased from BBInternational) through manipulation with an atomic force microscope (AFM) [23]. The corresponding experimental setup is shown in Fig. 2(a). A homemade inverted confocal microscope allows us to simultaneously perform optical measurements and nanoscale manipulation on particles. For excitation we use a frequency-doubled Nd:YAG laser (λ_{exc}=532 nm) and a frequency-doubled, picosecond-pulsed amplified diode laser (λ_{exc}=531 nm, 100 ps pulse width, 2.5 MHz repetition rate) allowing continuous wave and pulsed excitation, respectively. The polarization direction of the excitation light is controlled by a $\lambda/2$ waveplate. An oil immersion objective (60 × /1.4 NA) is used for focusing the excitation light onto the sample and for collection of the fluorescence emission. After passing a 550 nm longpass filter and a 50 μm pinhole, the fluorescence light can either be monitored by a CCD camera, spectrally dispersed and imaged by a spectrograph, or it can be analyzed in an HBT correlator (see inset in Fig. 1(b)). An AFM in tapping mode is used for mapping the topography of the sample, while manipulation of the particles is performed in contact mode. In a preliminary step the sample is prepared by spin-coating an aqueous solution of diamond nanocrystals and gold nanospheres onto a glass coverslip. Approximately 1% of the nanodiamonds contain a single NV center, as determined by comparing AFM measurements with corresponding confocal scans of the sample and analyzing the $g^{(2)}(\tau)$ function of the fluorescence from individual nanocrystals. These nanocrystals are then selected for subsequent experiments.

The assembly of the hybrid structure is performed by maneuvering gold nanospheres to a particular diamond nanocrystal via AFM manipulation and bringing all constituents into direct contact. In Fig. 2(b) two specific configurations are shown, namely a nanodiamond with one gold sphere attached (denoted as configuration A) and the same nanodiamond sandwiched between two gold spheres (denoted as configuration B). Three-dimensional finite-difference time-domain calculations are performed to estimate the plasmonic enhancement effects in these configurations (Fig. 2(c)). Thereby, the diamond nanocrystal is modeled as a truncated four-sided pyramid. Realistic material parameters are used and the influence of the glass substrate as well as the excitation with a high-NA objective is taken into account. Simulations of the excitation field predict a strong field enhancement only for polarizations along the x axis. In this case, pronounced hot spots are formed at the contact points between the diamond and gold particles, giving rise to an enhanced electric field inside the diamond nanocrystal, even if the latter is considerably smaller than the gold spheres. This effect leads to an enhancement of the excitation as well as emission rate of the NV center, provided that the dipole moment of the latter has a significant component in x direction.

From time-resolved measurements (Fig. 3(a)) on the configurations A (blue) and B (red) we obtain an increase in the excited-state decay rate $1/\tau_{dec}$ by a factor of 7.5 and 9.5, respectively. To check whether this enhancement is due to radiative or non-radiative processes, power-dependent measurements were performed (Fig. 3(b)). The data was fitted with a saturation model [40] of the form $P=\xi\sigma\Phi I_{exc}/(1+\sigma\Phi\tau_{rad}I_{exc})$, where σ is the absorption cross section, τ_{rad} is the radiative lifetime, Φ is the internal quantum efficiency, I_{exc} is the excitation intensity, and ξ is the total collection efficiency of our setup. Under strong excitation, the maximum number of emitted photons is only restricted by the radiative lifetime and the formula reduces to $P=\xi/\tau_{rad}$. Since our simulations show only a negligible influence of the gold spheres on the collection efficiency, we assume ξ to be constant and can thus deduce an increase in the radiative decay rate $1/\tau_{rad}$ by a factor of 5.8 and 8.9 for configurations A and B, respectively. This corresponds to quantum efficiencies $\Phi=\tau_{dec}/\tau_{rad}$ of 0.78 and 0.93, when $\Phi=0.99$ is assumed for the bare nanodiamond [41]. Obviously, in our configuration the first gold sphere induces a noticeable nonradiative decay channel (thus reducing the quantum efficiency), while the second gold sphere predominantly enhances the radiative decay rate (thus partly restoring the original quantum efficiency). This can be attributed to different distances of the gold spheres to the NV center and thus to a different impact of fluorescence quenching effects [16,17]. Note that the enhancement of the radiative decay rate by nearly an order of magnitude is, to our knowledge, so far the highest achieved for NV centers in