Alignment of plasmonic nanostructures to self-assembled quantum dots with sub-10 nm accuracy

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Controlling light-matter interaction of single quantum emitters by engineered nanostructures is a highly topical area of quantum optics. Developments in fabrication methods have allowed realizing, e.g., optical nonlinearities on the single-photon level for quantum dots coupled to photonic crystal cavities, enhanced spontaneous emission into free space using plasmonic structures, and unidirectional single photon emission from an optical antenna excited by a single quantum dot. The quality of the control depends critically on the ability to position and orient the nanostructures with respect to the quantum system. For plasmonic structures the required accuracy is on the nanometer scale, as plasmonic fields decay on a length scale much shorter than the wavelength. Figure 1a shows the calculated enhancement of the electric field around a plasmonic nanoantenna. Around such a nanostructure the electric field is focused into hot spots on a 10 nm length scale. Here we demonstrate a highly flexible method that allows fabricating plasmonic nanostructures aligned to stable solid-state quantum emitters with a precision better than 10 nm in order to control the interaction of optical fields with single quantum systems.

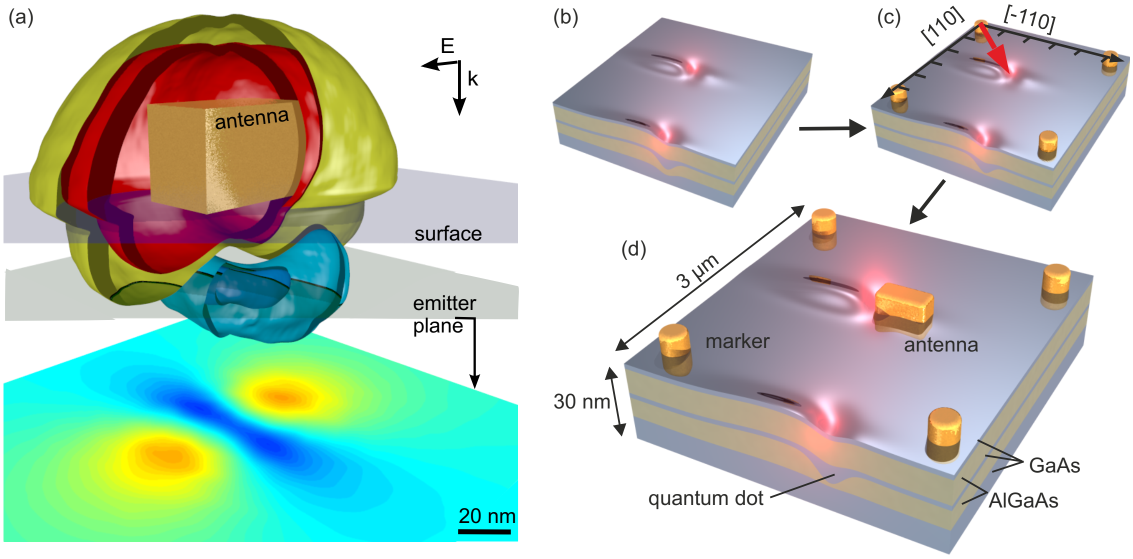

Figure 1: The strong localization of the optical field near plasmonic nanostructures requires an excellent positioning accuracy to obtain reliable coupling between quantum emitter and plasmonic nanostructure. (a) Electric field enhancement around a plasmon-resonant gold antenna with dimensions $20 \times 35 \times 20$ nm^3 on a substrate with refractive index 3.5. The isosurfaces correspond to contours where the field amplitude is 1.5 and 2 times higher or lower than in the absence of the antenna. The pseudo-color plot in the bottom plane displays the field 20 nm beneath the sample surface where single quantum dots can be located. *Right:* Main steps of the nanofabrication method to position plasmonic structures with respect to quantum dots with 10 nm accuracy: (b) The position of GaAs quantum dots can be determined from a characteristic topography feature. (c) We use a grid of gold markers fabricated with electron beam lithography to define a coordinate system on the sample in order to determine the positions of quantum dots. (d) A plasmonic nanoantenna is positioned next to the target quantum dot in a second electron beam lithography step. Here, quantum dot and antenna are drawn to scale but the markers are further apart.

Our fabrication method is schematically illustrated in the right part of Fig. 1. It combines single solid state quantum dots whose position can be determined with high accuracy, and a nanofabrication process for multiple aligned layers. The quantum dots are self-assembled GaAs dots in AlGaAs barriers. The samples are grown using molecular beam epitaxy. The quantum dot has two almost energetically degenerate exciton transitions with mutually orthogonal transition dipole moments with fixed orientation with respect to the crystal axes. More importantly, above each quantum dot we find a characteristic topography feature that signals the position of the emitter (see Fig. 1b) [1]. The surface feature allows us determining the position of quantum dots with nanometer accuracy using high-resolution microscopies such as scanning electron microscopy (SEM) or atomic force microscopy (AFM). The quantum dots are located in a layer 18 nm away from the interface so that they can be coupled to plasmonic structures fabricated on the sample surface. Our nanofabrication process consists of first fabricating a set of markers using electron beam lithography. The orientation of the marker grid is aligned with the crystal axes. The position of a quantum dot can be determined in the coordinate system defined by the markers using pattern matching (Fig. 1c). Finally, the coordinates of the target quantum dot can be used to fabricate the desired nanostructure in a second electron beam lithography step. In the case shown in Fig. 1d an optical antenna is fabricated in this way in order to enhance the emission from the quantum dot. Our fabrication method allows positioning nanostructures with respect to the quantum dots with nanometer scale accuracy and oriented precisely with respect to the transition dipole moments.

To determine the precision of our fabrication technique we prepare a sample with gold nanorod antennas positioned next to single quantum dots. Each antenna is fabricated so that one end is centered on the elliptical topography feature related to the quantum dot. The in-plane dimensions of the gold nanorods are about 22 nm $\times$ 56 nm which allows us to determine the position of the antenna with nanometer scale precision. High-resolution scanning electron micrographs of quantum dots with and without a plasmonic antenna are shown in Fig. 2a. A high electron dose in SEM imaging results in significant degradation of the optical properties of the quantum dots. We therefore reduce the integration time when determining the position of single quantum dots to avoid sample degradation and to also reduce sample drift and contamination. Typical examples of the raw images used for position determination are displayed in the second row of Fig. 2a. We reduce noise in the micrographs by low-pass and Gaussian smoothing filters (Fig. 2b). To obtain the positions, we place elliptical and rectangular frames with fixed dimensions on the quantum dot and antenna, respectively. The positions that best match the filtered electron micrographs are determined by eye. Small displacements from the optimal position are clearly visible by eye as illustrated in Fig. 2b. Here the frames have been displaced from the optimum by 10 nm in both the horizontal and vertical directions. Compared to a numerical cross-correlation method our manual method was found to be more robust and as sensitive. From the best-fit positions the mid-point of the ellipse and the end-point of the rectangle are used to define the positions of the quantum dot and optical antenna (marked by a cross and a circle, respectively).

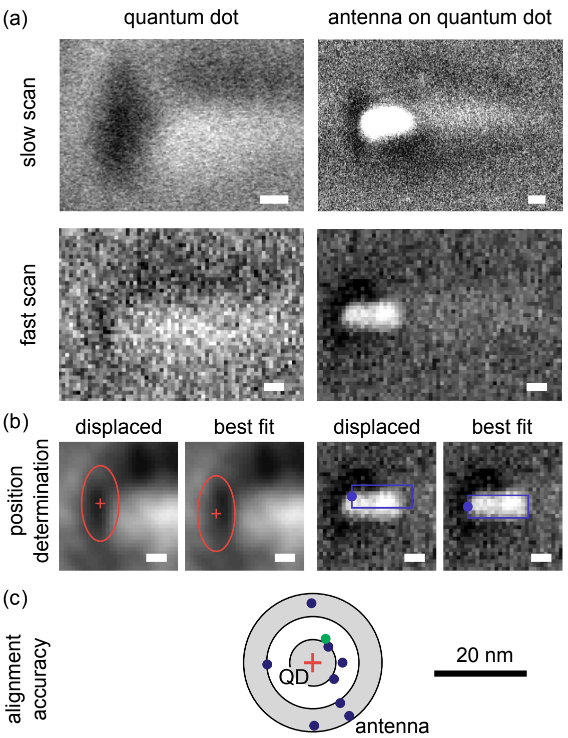

Figure 2: Positioning a plasmonic nanoantenna on a quantum dot: (a) High resolution electron micrographs using long exposure times clearly show the topography feature of the quantum dot and the gold antenna. In order to avoid sample contamination and degradation as well as sample drift, fast-scan, low resolution images have to be used for the position determination. (b) Frames in the shape of ellipses and rectangles are used to determine the position of the quantum dot and antenna, respectively. The best-fit position is determined by eye using a filtered electron micrograph with reduced noise. The accuracy of the manual position-determination method is illustrated by displacing the frames by 10 nm in horizontal and vertical directions away from the best fitting position. (c) Distribution of the positions of nanoantennas with respect to the target quantum dots. The quantum dot selected for TEM imaging is marked with a green circle. All scale bars are 20 nm.

Using the approach described above, the positions of selected quantum dots were determined in the coordinate system defined by the marker grid. Nanorod antennas were then fabricated aligned to the target quantum dots in a second electron beam lithography step. Finally, the positions of the realized antennas were determined in the coordinate system defined by the marker grid. The spread in the position of the realized optical antennas with respect to their target quantum dot is shown in Fig. 2c. The average distance between antenna and emitter is 9.3 nm illustrating the accuracy of our approach.

To verify the position of the quantum dot with respect to the surface topography feature we prepare a thin lamella containing a single quantum dot decorated with an optical antenna for cross-section transmission electron microscopy (TEM). A scanning electron micrograph of the structure chosen for the TEM imaging is shown in Fig. 2b. A dark-field TEM micrograph of the lamella is displayed in Fig. 3. The quantum dot is seen to be perfectly located below the dip in the sample surface. Furthermore, the plasmonic antenna is exactly at the design position. The difference between the position of the quantum dot and nanorod is only 2.2 nm as determined from the centers of mass of cross-sections passing through the emitter and antenna. As well as illustrating the excellent accuracy of our positioning method and its implications for realizing advanced plasmonic structures, Fig. 3 is the first TEM micrograph of a strain-free GaAs/AlGaAs quantum dot. Our positioning method allows marking preselected nanostructures for high-resolution TEM imaging in order to correlate data from optical experiments with structural information down to atomic resolution.

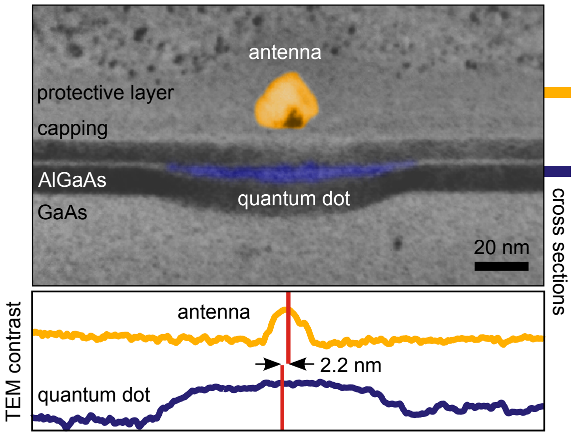

Figure 3: *Upper panel:* cross-section transmission electron micrograph (inverted dark field) of optical antenna (yellow) positioned above a single GaAs quantum dot (blue). The quantum dot is the thicker part of a thin GaAs quantum well (bright) between AlGaAs barrier layers (dark). *Lower panel:* cross sections across the metallic structure and the quantum dot at positions indicated by the colored bars beside the micrograph in the upper panel. The cross sections show that the gold nanostructure is almost perfectly centered on the quantum dot with the centers of mass of the cross sections, indicated by red lines, differing by only 2.2 nm.

We have demonstrated a method to position nanostructures aligned with sub-10 nm precision with respect to single GaAs semiconductor quantum dots. The achieved positioning accuracy is better than the characteristic length scales of optical fields around plasmonic structures paving the way to realizing advanced plasmonic nanocircuits containing solid-state quantum emitters. Our fabrication technique is highly flexible allowing the nanostructures to be prepared using state-of-the-art methods such as electron beam lithography and focused ion beam milling. As a first application of the developed positioning technique we were able to acquire the first TEM micrograph of a low-density self-assembled GaAs/AlGaAs quantum dot.

References:

[1] *Pfeiffer, M., K. Lindfors, C. Wolpert, P. Atkinson, M. Benyoucef, A. Rastelli, O.G. Schmidt, H. Giessen, and M. Lippitz.* Nano Letters **10**, 4555 (2010).

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