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Efficient coupling of single organic molecules to channel plasmon polaritons supported by V-grooves in monocrystalline gold

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Abstract

Single dibenzoterrylene (DBT) molecules in anthracene nanocrystals have been shown to emit single photons without blinking, bleaching or spectral diffusion. Efficient integration of these molecules into photonic circuits will pave the way toward the realization of quantum optical networks. Here, we incorporate anthracene nanocrystals containing single DBT molecules into plasmonic V-grooves milled with a focused ion beam in monocrystalline gold flakes. The fabricated V-grooves exhibit high-quality low-loss guiding of channel plasmon polaritons with the propagation length reaching
-14 μm at a wavelength of 800 nm. For DBT molecules coupled to the V-grooves, we observe enhanced emission decay rates with up to 50% of the emission being funneled into channel plasmon polaritons.

Keywords

Quantum Plasmonics, Quantum Optics, V-groove, Dibenzoterrylene (DBT) molecule, Gold Flake

Single photon emitters with suitable properties are required for quantum and communication technologies. Various single photon emitters are explored for its suitability in quantum technologies. One of the promising candidates for its application as a single photon emitter is dibenzoterrylene (DBT) molecules embedded in anthracene (Ac) crystals. DBT-Ac crystals have been shown to be stable single photon emitters with properties such as indistinguishable photon emission, high quantum efficiency and stable emission wavelength. These properties were utilized for Hong-Ou Mandel interference experiments and DBT molecules were found to emit lifetime-limited indistinguishable photons at cryogenic temperatures. The emission from DBT molecules has been modified using fiber microcavities to correspond to a two-level system. Recently, it has been shown that DBT molecules embedded in nanocrystals retain their lifetime-limited linewidths and spectral stability. Moreover, the emission from DBT molecules has been controlled electrically by integrating them with 2D materials. These properties of DBT molecules in Ac-nanocrystals make them suitable for application in quantum information technologies. Efficient integration of these molecules into waveguides will pave the way for their usage in quantum optical networks.

Efficient coupling between a quantum emitter and a waveguide can be achieved if the waveguide supports a very confined mode. DBT molecules have been coupled to dielectric waveguides, and the efficiency of coupling to waveguides has been limited to 42%, as the mode confinement is limited. On the other hand, plasmonic waveguides can support very confined modes and have been utilized for coupling of various emitters. For DBT
molecules, it has been theoretically predicted that the coupling to a plasmonic slot mode can be up to 97%.\textsuperscript{30} Plasmonic V-grooves are promising waveguide structures for coupling to single emitters, as it provides with a combination where the mode is confined and the propagation length is relatively long.\textsuperscript{21,31,32} Moreover, many components of an optical network such as beam-splitters, ring resonators have been demonstrated.\textsuperscript{33} With a tapered V-groove structure, regions of highly confined modes can be combined with regions of longer propagation lengths.\textsuperscript{34,35} Here, we utilize plasmonic V-grooves fabricated in monocrystalline gold flakes by milling with a focused ion beam (FIB) of gallium ions and subsequently polished with helium ions. This resulted in low-loss plasmonic waveguides supporting the propagation of channel plasmon polaritons (CPPs) featuring the propagation lengths of \textasciitilde14 \mu m at a wavelength of 800 nm. We further study experimentally DBT-Ac nanocrystals inserted in V-grooves and observed that DBT molecules couple well to CPPs, funnelling up to 50% of their emission into the CPP mode.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure1.png}
\caption{Schematic of the experiment. An anthracene nanocrystal containing a single DBT molecule is excited by 730 nm laser, the fluorescence from DBT molecule excite single plasmons in V-groove. The plasmons propagate to the end of V-grooves and get out-coupled to far-field by nano mirrors. A small portion of the fluorescence is emitted directly to the far-field.}
\end{figure}

In our experiments, single DBT molecules embedded in Ac nanocrystals are inserted in FIB-milled V-grooves in a monocrystalline gold flake and excited by a 730 nm laser (Figure 1). Excited single DBT molecules generate single CPPs that propagate along the plasmonic V-grooves away from the molecules, being eventually outcoupled by nano-mirrors.
fabricated at the V-groove terminations.\textsuperscript{36} Part of the fluorescence from DBT molecules is directly emitted into the far-field as photons.

Gold monocrystalline flakes were prepared using air-thermolysis of a precursor, in which (AuCl$_4$)$^-$ phase transferred from aqueous medium and stabilized in toluene by tetraoctylammonium bromide (ToABr), to yield large hexagonal and triangular Au microplates.\textsuperscript{37–39} More details of the gold flake preparation can be found in the supplementary information (SI). We utilized a two-step focused ion beam lithography process to fabricate V-grooves with defined dimensions, smooth side-walls and inclined end-faces on monocrystalline gold flakes. The subsequent application of gallium and helium focused ion beams (GaFIB/HeFIB) provided by a commercial instrument (Zeiss Orion Nanofab) allowed us to fabricate waveguides of the required size and shape at reasonable fabrication speed.

First, we used GaFIB to mill V-grooves of different lengths (10 $\mu$m, 12.5 $\mu$m, and 15 $\mu$m) with a central width $w = 180 \pm 10$ nm, an opening angle of 14.5° and a depth $d = 700 \pm 100$ nm. We varied the dose across the groove to control the opening angle. The structures have been made with a maximum dose of 0.36 nC/$\mu$m$^2$ at the edges that was reduced to 0.18 nC/$\mu$m$^2$ towards the center of the structure. In Figure 2, we present optical microscope and helium ion microscope (HIM) images of V-grooves fabricated by FIB milling of a gold flake. In order to facilitate efficient coupling of free-propagating light to CPPs propagating in V-grooves, we applied a linear dose gradient at the upper and lower end of the V-groove to create a funnel-like end-face (see Figure 2(c) and (d)). We finalized the V-grooves by HeFIB using a constant helium ion dose of 0.05 nC/$\mu$m$^2$. After this process, we found smooth structures of the desired size and shape without contamination by gallium ions.

V-grooves fabricated in this manner support the propagation of a well-confined CPP mode (Figure 3(a)). The mode field distribution is calculated with the finite element method (FEM) using commercial software (COMSOL Multiphysics). V-grooves can support extremely confined and lossy modes as well.\textsuperscript{40} These modes, in our case, are not excited by DBT molecules due to the relatively large size of our nanocrystals. Next, we optically charac-
Figure 2: (a) Microscope image of a gold flake containing V-grooves of different sizes. (b) HIM image of V-grooves. (c) HIM image of a cross-section of a V-groove together with a nano mirror. (d) HIM image of the nano-mirror at one of V-groove ends.

Figure 3: (a) Electric field of a waveguide mode supported by a V-groove with height h=700 nm and width w=180 nm. In the simulations, corners are rounded with a radius of 10 nm. Arrows indicate the direction of the electric field at their tails and their lengths are proportional to the magnitude of the electric field at the tail. (b) V-grooves of different lengths are excited by a supercontinuum laser filtered around 800 nm. Lengths of V-grooves are noted in different panels. The out-coupled spots are indicated by arrows. (c) Measured propagation lengths as a function of wavelength are plotted. The error-bars are one standard deviation for the experiments.
terized V-grooves to obtain their propagation losses as a function of wavelengths. We shone supercontinuum laser (SuperK, NKT Photonics) filtered at different wavelengths at one end of the V-groove and took images, for different lengths of V-grooves (10 μm, 12.5 μm, and 15 μm). A set of images taken with excitation at a wavelength of 800 nm are presented in Figure 3(b) (For wavelengths 700 nm, 750 nm, 850 nm and 900 nm, images can be found in the SI). A reflection image is also taken using suitable neutral density (ND) filters so that the reflection images are not saturated. These images are used to obtain the total insertion loss for different V-grooves. By fitting total insertion loss for different lengths of V-grooves, we obtained propagation losses. In Figure 3(c), we present measured propagation lengths as a function of wavelengths for six sets of V-grooves. As expected, propagation lengths increase as a function of wavelength. At 800 nm, we obtain a propagation length of 13.6 ± 1.9 μm. Experimentally measured propagation lengths are longer than propagation lengths obtained in simulations (see SI), where refractive indices from Johnson and Christy are used. The propagation length obtained in simulations at 800 nm wavelength is 9.4 ± 1.5 μm. This is attributed to the monocrystalline nature of gold flakes utilized for the fabrication of V-grooves, as well as the smoothness of V-grooves obtained due to the fabrication process, described earlier in this letter. A similar effect has been observed for the propagation of surface plasmon polaritons on monocrystalline silver flakes, where propagation lengths of SPPs on monocrystalline silver flakes exceeded that obtained in simulations with Johnson and Christy data.

Having characterized the V-grooves, we now present the fabrication process of anthracene nanocrystals containing single DBT molecules and their characterization. To fabricate the nanocrystals, we have used a slightly modified version of the procedure used by S. Pazzagli, et al. The crystals are fabricated using a reprecipitation method. In this procedure, a dilute solution of anthracene was prepared with a water-soluble solvent (acetone, in our case) and DBT molecules are dissolved in toluene. The desired amount of DBT-toluene solution is mixed with Ac-acetone solution. When Ac-acetone containing DBT molecules is injected
Figure 4: (a) AFM topography image of anthracene nanocrystals doped with DBT molecules. (b) Fluorescence image of anthracene nanocrystals containing DBT molecules. (c) Typical spectrum obtained for single DBT molecules in Ac-crystals. (d) Typical auto-correlation obtained for single DBT molecules in Ac-crystals. (e) Lifetime of a single DBT molecule, fitted with a single exponential. (f) Histogram of DBT-molecule lifetimes.
into sonicated water, it divides into many droplets. The solvent gradually dissolves, and correspondingly the concentration in the microdroplets becomes supersaturated until the anthracene, which instead is not water-soluble, is reprecipitated in the form of nanocrystals. More details of the procedure can be found in SI.

In Figure 4, we present the results of the characterization of Ac-crystals embedded with DBT molecules. Figure 4(a) shows an atomic force microscope (AFM) image of the crystals, which shows that the maximum height of these crystals is 70 nm. Figure 4(b) shows a fluorescence image taken at an excitation power of 50 μW at a pulse rate of 10 MHz. Detailed schematic of our optical set-up utilized for characterization of DBT molecules as well as DBT molecule-V-groove coupled system can be found in SI. Typical fluorescence obtained from these crystals is presented in Figure 4(c). In Figure 4(d), we present a typical auto-correlation measured for single DBT-molecules. Measured data as well as a single exponential fit to the data is presented, which is for the highest obtained lifetime for DBT-molecules in Ac-crystals. In Figure 4(f), we present a histogram obtained for lifetimes of DBT molecules in anthracene crystals. The average lifetime for DBT molecules is 4.49 ± 0.74 ns, where 0.74 ns is one standard deviation. We observe that the lifetime distribution is broader compared to what was observed before. This could be due to smaller size and wider size distribution, resulting in lower local optical density of states inside these small crystals.

Having obtained desirable V-grooves as well as DBT-Ac crystals, we coupled them by spin coating the DBT-Ac crystals on samples containing V-grooves. We obtained many coupled systems by following this procedure. One such coupled system is presented in Figure 5. Dark-field microscope images comparing the V-grooves before and after spin-coating of DBT-Ac nanocrystals can be found in SI. In Figure 5(a), we show the fluorescence image when DBT-molecule is excited by 730 nm laser light. In addition to the direct spot associated with DBT, two additional spots corresponding to the ends of the V-groove (A and B) can be seen in the image. This is due to the excitation of CPPs in the V-groove by the DBT molecule, subsequent propagation of CPPs to the ends of the V-groove and scattering of CPPs to
Figure 5: (a) Fluorescence camera image while DBT molecule is excited. Two extra spots appear due to excitation of plasmons by DBT-molecules in V-grooves. Plasmons in V-grooves propagate to the ends of V-groove and get scattered to the far-field (Spots A and B). (b) Emission spectrum for the spot DBT. (c), (d) and (e) Lifetime measurement data and a single exponential fit to the data for spots DBT, A and B, respectively, indicated in (a). (f) auto-correlation measurement for spot DBT. (g) and (h) cross-correlation measurement between spots DBT, A and DBT, B, respectively.
the far-field by nano mirrors at the ends of the V-groove. In some cases, small nanocrystals containing no DBT molecule can also go into the V-groove and can scatter, which gives rise to more spots observed. This, however, is not the case in most of the systems. In Figure 5(b), we present the spectrum obtained from the DBT spot in Figure 5(a). This is very similar to the typical spectra for DBT molecules at room temperature. In Figure 5(c), we present data and a single exponential fit to the lifetime measurement data for the DBT spot. The lifetime obtained is slightly lower than the mean lifetime obtained for DBT-Ac crystals on a glass substrate. However, a direct comparison for change in decay-rate cannot be made. In Figure 5(d) and (e), we present the lifetimes measured for spots A and B, which are very close to the lifetime obtained for the DBT spot and are within the error margin ($\pm 0.1$ ns). This is a clear indicator of the spots DBT, A and B originating from the same single molecule. In Figure 5(d), we present the auto-correlation measurement for the DBT spot, where $g^2(0) < 0.5$ clearly suggests that the nanocrystal coupled to the V-groove contains a single DBT molecule. We also present cross-correlation measured between spots DBT, A and DBT, B in Figure 5(d) and (e), respectively. Again, $g^2_{DBT,A}(0) < 0.5$ and $g^2_{DBT,B}(0) \approx 0.5$ clearly suggest that spots DBT, A and B originates from the same single DBT molecule. We note that $g^2_{DBT,A}(0) \approx 0.5$ due to lower signal-to-noise ratio for spot B. The lower signal-to-noise can arise from Au fluorescence, which can inefficiently couple to the V-groove mode.\cite{21}

$\beta$-factor is an important figure-of-merit for emitter-waveguide coupled systems, defined as the ratio of decay-rate into the waveguide mode and total decay-rate. From the fluorescence image in Figure 5(a), we estimate the $\beta$-factor as $\beta = (I'_A + I'_B)/(I'_A + I'_B + I_{DBT})$, with $I'_A = I_{AC}(L_A/L_p)/\eta_{VTM}$, $I'_B = I_{BE}(L_B/L_p)/\eta_{VTM}$ and $I'_{DBT} = I_{DBT}/\eta_{DBT}$ where $I_{DBT}$, $I_A$ and $I_B$ are the intensities measured at spots DBT, A and B, respectively. $L_A$ and $L_B$ are distances of ends A and B from DBT, respectively. $L_p$ is the propagation length for the V-groove waveguide mode. We use the value obtained before for propagation length (14 $\mu m$). $\eta_{DBT}$ and $\eta_{VTM}$ are collection efficiencies at DBT and V-groove ends respectively.
With numerical simulations, we obtain $\eta_{DBT}$ and $\eta_{VTM}$ to be 68% and 70%, respectively (see SI for details about the dependence on position and orientation of dipole on collection efficiency). We estimate a $\beta$-factor of 31% for the coupled system presented above. We note that we have neglected non-radiative decay in the $\beta$-factor estimation, which is reasonable as the lifetime change is less than a factor of 2 for the coupled system.

Figure 6: (a) A schematic showing the coupled system together with axes for reference. (b-e) Polarization analysis of a coupled system. Green arrows show the excitation laser polarization. Red arrows indicate the analyzer polarization of fluorescence. From top to bottom, the analyzer polarizations are (b) perpendicular to V-groove axis, (c) 45° to V-groove axis, (d) along V-groove axis, and (e) 135° to V-groove axis.

We have observed more coupled systems for which, we measured decay rates and emission from the distal ends. In table 1, we summarize the properties of 7 coupled systems. System 1 is presented in the main text of this manuscript, and fluorescence images of 6 other systems can be found in the SI. We note that DBT-Ac can bleach out in experiments at room temperature. In our experiments, it took 1-4 hours for them to bleach out. For all the systems, presented in table 1, fluorescence image, lifetime, auto-correlation and spectrum were measured.

We have studied polarization properties of coupled systems to better understand the
Figure 7: β-factor and $\Gamma_{tot}/\Gamma_0$ as a function of position along the line indicated in the insets, for (a) dipole along x-axis and (b) dipole oriented 45° to x-axis in xz-plane. Grey band shows the region where a nanocrystal of size 70 nm would fit in the V-groove. Insets in the graphs present decay-rate in the CPPs normalized to decay-rate in vacuum for corresponding orientations of dipole.

coupling between DBT-Ac and V-grooves. Figure 6 shows polarization-dependent fluorescence from a coupled system. All the four images in the figure are obtained with the same excitation polarization and power. It is clear from Figure 6 that the emission from V-groove ends are maximum when the analyzer polarization is perpendicular to the V-groove axis. Also, when the analyzer polarization is along the V-groove axis, emission from the V-groove ends can not be observed, which clearly suggests that the V-groove mode is excited by the DBT molecule. Also, the emission from DBT spot is brightest when the analyzer position is 45° to the V-groove axis, and dimmest when the analyzer is 135° to the V-groove axis. This suggest that the dipole orientation is 45° to the V-groove axis. There is emission into the V-groove mode as long as there is a finite projection of the dipole along the electric field of the mode as shown in Figure 3(a).

Table 1: Properties of seven coupled systems.

<table>
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<th>Sl. No.</th>
<th>1</th>
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<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
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</thead>
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<td>Lifetime of coupled DBT molecules (ns)</td>
<td>4.0</td>
<td>4.0</td>
<td>4.1</td>
<td>4.2</td>
<td>4.8</td>
<td>3.9</td>
<td>4.0</td>
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<tr>
<td>β-factor</td>
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<td>0.50</td>
<td>0.33</td>
<td>0.23</td>
<td>0.19</td>
<td>0.37</td>
<td>0.33</td>
</tr>
</tbody>
</table>

We now compare our experimental results with numerical simulations. In Figure 7(a)
and (b) insets, we present the decay-rate into the plasmonic mode ($\Gamma_{pl}$) normalized by its decay-rate in the vacuum, when the dipole is oriented along x-axis and 45° to x-axis in xz-plane [Figure 6(a)]\(^{13}\). We observe that the decay-rate into the plasmonic mode could be up to 20 times the decay-rate in the vacuum, if a nanocrystal is small enough to fit into the very bottom of the V-groove. For larger crystals, the decay-rate changes will naturally be lower. For example, for a 70 nm anthracene nanocrystal, the decay-rate into the plasmonic mode will be up to 6 times the decay-rate in the vacuum. For a dipole oriented 45° to V-groove axis, the decay-rate into the plasmonic mode ($\Gamma_{pl}$) normalized by its decay-rate in the vacuum is ~2 times lower compared to dipole oriented along x-axis. Furthermore, the very bottom of the V-groove does not represent the optimal position of the emitter with respect to maximizing the $\beta$-factor, as discussed below. Structural changes can also affect the decay-rate into the plasmonic mode. In SI, the simulation results due to structural changes in V-groove can be found.

To calculate the $\beta$-factor total decay-rate, we again followed the method of Chen et. al.\(^{13}\) We position point dipole emitting at 785 nm parallel to the electric field at different points along the line in Figure 7(a) inset in a 3D FEM model, and obtained total decay-rate normalized to its decay-rate in the vacuum ($\Gamma_{tot}/\Gamma_0$). Total decay-rate ($\Gamma_{tot}$) consists of decay-rate into CPPs($\Gamma_{pl}$), far-field ($\Gamma_{rad}$) and non-radiative($\Gamma_{nonrad}$) channels. The $\beta$-factor ($\Gamma_{pl}/\Gamma_{tot}$) and $\Gamma_{tot}/\Gamma_0$ are plotted in Figure 7(a) and (b) for dipole oriented along x-axis and 45° to x-axis in xz-plane. From Figure 7(a), we observe that a $\beta$-factor of up to 63% can be obtained. We note that largest $\beta$-factors cannot be obtained for emitters at the very bottom of the V-groove, because the decay-rate into the non-radiative modes increases drastically for small emitter separations from the metal surface occurring at the bottom.\(^{21}\) Instead, the largest $\beta$-factor is obtained at a height of 250 nm from the bottom. This also coincides with the area where an anthracene nanocrystal of size 70 nm can be positioned. Because of the random location of DBT molecules in nanocrystals, a $\beta$-factor in the range of 0.45 - 0.63 [grey band in Figure 7(a)] can be obtained for molecules with dipoles aligned.
along the electric field of the channel plasmon polariton mode. Random orientations of the transition dipole moment can further decrease the emission coupling to CPPs in V-grooves. In addition, it can also decrease the decay into non-radiative channels. In Figure 7(b), we observe that the maximum $\beta$-factor is 67\% for dipole oriented 45° to x-axis in xz-plane, even though the total decay rate is lower compared to dipole along x-axis at the same position.

In experiments, we have achieved a $\beta$-factor of 50\% in one case, and $\beta$-factors in the range of 19-37\% in other cases. The average lifetime for DBT molecules coupled to V-grooves is $4.1 \pm 0.3$ ns and the average $\beta$-factor is $0.32 \pm 0.10$. On average, the decay-rate enhancement amounts to 1.1. Considering the range of lifetimes for uncoupled DBT molecules and that of coupled molecules, the change in molecule lifetimes varies from 0.9 to 1.6. DBT molecules in small anthracene crystals will have longer lifetimes and they couple better to V-grooves. Therefore, the lifetime changes for the well-coupled system is found towards the upper limit in the aforementioned range. Furthermore, in experiments the lifetimes are compared between DBT-Ac in V-grooves to that on a fused silica substrate. The substrate decreases the lifetime of the emitter compared to the emitter placed in vacuum.\textsuperscript{45} With all the factors - random position of emitter inside the nanocrystal, orientation of dipole and substrate - the expected lifetime changes correspond closely to the lifetime changes obtained in the experiments.

We now compare our experimental results with other waveguide-DBT coupled systems. For single DBT molecules coupled to silicon nitride waveguides, the maximum coupling was observed to be 42\% with the average coupling efficiency amounting to only 20\%.\textsuperscript{14} For other dielectric waveguide-DBT molecule coupled systems,\textsuperscript{15} the coupling efficiency reported is even less than 20\%. Coupling of DBT molecules to hybrid plasmonic gap waveguide modes has also been reported,\textsuperscript{29} with coupling efficiency reaching only 11.6\%. Therefore, we note that the $\beta$-factor achieved in our experiments is larger than those reported in the literature for DBT-molecules coupled to dielectric or plasmonic waveguides.

In conclusion, we have realized coupling between single DBT molecules in anthracene nanocrystals and CPPs propagating in V-grooves milled in monocrystalline gold. Long prop-
agation lengths for CPPs in V-grooves is obtained due to a combination of monocrystalline flakes and two-step focused ion beam milling to obtain the V-grooves. Ga ion beam was used to fabricate the V-grooves, whereas He ion beam was used to polish the surface and remove the surface contamination. With the placement of nanoparticles inside the V-grooves, we observe the CPP excitation efficiency ($\beta$-factor) of up to 50%. As an outlook, with smaller Ac-crystals, deterministic positioning techniques used in combination with plasmonic waveguides supporting highly confined modes, efficiently coupled DBT-waveguide systems can be obtained making prospects of quantum optical networks realistic.

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Supporting Information Available

S1: Growth of gold flakes, S2: Growth of anthracene nanocrystals embedded with dibenzoterrylene molecules, S3: Optical set-up, S4: V-groove images, S5: Coupled Systems, S6: Simulations

References


Graphical TOC Entry

![Diagram](image_url)