An optical nanofiber–based interface for single molecules

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Optical interfaces for quantum emitters are a prerequisite for implementing quantum networks. Here, we couple single molecules to the guided modes of an optical nanofiber. The molecules are embedded within a crystal that provides photostability and, due to the inhomogeneous broadening, a means to spectrally address single molecules. Single molecules are excited and detected solely via the nanofiber interface without the requirement of additional optical access. In this way, we realize a fully fiber–integrated system that is scalable and may become a versatile constituent for quantum hybrid systems.

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In recent years, single molecules in solids 1–7 and other solid state quantum emitters such as color centers in diamond 8–11 and quantum dots 12–16 have gained increasing interest as building blocks for quantum networks 17–18, quantum metrology 19–21 and nanosensors 22–24. For all these applications a strong light–matter interaction is essential. This can be achieved by coupling to a large ensemble of quantum emitters 25–26, by employing a cavity 27–30 or by decreasing the mode area of the interacting light field 31–32 and hence achieving a significant overlap between the absorption cross-section of the emitter and the respective light field. A versatile platform to achieve such a small mode area of the light field are optical nanofibers 33–41. An optical nanofiber is the waist of a tapered optical fiber (TOF) and has a diameter smaller than the wavelength of the light it is guiding. Therefore, an appreciable fraction of the light propagates outside the fiber in the form of an evanescent wave. Due to the strong transverse confinement of the light field, which prevails over the entire length of the nanofiber, the interaction with emitters close to the surface can be significant 33–35.

Single molecules in crystalline solids are efficient quantum emitters that exhibit strong zero phonon lines (ZPL) which can be lifetime-limited and as narrow as tens of MHz at cryogenic temperatures 42–43. Due to inhomogeneous broadening caused by the host crystal such molecules can be spectrally discerned and individually addressed using a narrowband laser 44. For a low concentration of molecules, this makes it possible to circumvent the additional spatial selection that has been used for numerous single molecule experiments in the past 45–46, even if a large fraction of the crystal is illuminated. This was also exploited in recent experiments 31–32, where single dibenzoterrylene molecules have been coupled to light propagating through a nanocapillary. Single molecules come in a large variety and they are small quantum emitters which is useful when coupling them to nano- and microcavities 48–50 and also offers the possibility to study collective phenomena of quantum emitters. Additionally, single molecules such as polycyclic aromatic hydrocarbons can be spectrally very stable and do not suffer from photobleaching when embedded in the right host matrix. In addition to a near-unity quantum yield, these are very important features when working with solid state emitters. Here, we show for the first time that single organic molecules can be interfaced with an optical nanofiber. This presents a new platform based on solid state emitters that can be used for quantum optics and that is naturally integrated into optical fiber networks.

In our experimental set-up, the TOF resides inside a cryostat [Fig. 1(a)] and we interface terrylene molecules in a para–terphenyl (p–terphenyl) crystal with the evanescent light field surrounding its nanofiber. The latter has a total length of 3 mm and a diameter of 320 nm. The TOF is produced in a heat and pull process using a custom–made pulling rig 51. In the 6.8 cm long tapered section of the fiber, the weakly guided LP01 mode of the standard single mode optical fiber is adiabatically transformed into the strongly guided HE11 mode of the nanofiber waist and back yielding transmission losses of less than 2% from 520–650 nm. For our purpose, a broadband transmission is crucial as the excitation and detection wavelengths can differ by more than 100 nm. This requires a careful choice of tapering angles and waist diameter 52. Terrylene in p–terphenyl can exhibit four different electronic transition frequencies from the ground to the first excited state termed X1–X4, corresponding to four possible orientations of the molecules in the crystal. Molecules in the X4 orientation are resonant with light at 577.9 nm and have been shown to be very photostable 53. Hence, all measurements presented here use molecules in this site. A simplified level diagram is depicted in Fig. 1(b). The laser excites the molecule on the zero phonon line 00ZPL that connects the ground and the excited electronic state.
without any vibrational contribution of the molecule. After excitation, the molecule will decay into any of the vibrational states in the electronic ground state with a probability determined by the Franck-Condon $\alpha_{FC}$ and Debye-Waller $\alpha_{DW}$ factors. From these states it will non-radiatively decay into the vibrational ground state within picoseconds. Hence, the absorption cross-section \[ \sigma = \alpha_{FC} \alpha_{DW} \frac{3\lambda^2}{2\pi} \frac{\Gamma^2}{\Gamma_{hom}^2} (\hat{d} \cdot \hat{e})^2 \] of a single molecule in a solid is where the product $\hat{d} \cdot \hat{e}$ represents the projection of the polarization vector $\hat{e}$ of the excitation light on the unit vector of the molecular dipole $\hat{d}$ and $\lambda$ denotes the excitation wavelength. $\Gamma$ is the lifetime-limited linewidth and $\Gamma_{hom}$ the homogeneously broadened linewidth of the 002ZPL, respectively. If the molecular dipole is aligned with the polarization of the excitation light and the homogeneous broadening is negligible, the absorption cross-section will be that of a simple two-level atom and is comparable to the effective mode area $A_{eff}$ \[ \sigma \] of our optical nanofiber of about $0.5 \times \lambda^2$. This ensures a strong effect of a single molecule on the light field. The probability of a terrylene molecule to decay to the triplet state after excitation rather than to the singlet ground state is very low and has experimentally been found to be $< 10^{-5}$ at cryogenic temperatures \[ [2, 56] \]. To maintain sufficient light guiding capabilities of the nanofiber-crystal system for spectroscopy and to ensure that the crystal stays tightly adhered to the vertically mounted nanofiber, the crystals have to be on the order of a few hundred nanometers in size. Such nanocrystals are grown by a reprecipitation method \[ [59] \] from an oversaturated solution of a $8 \times 10^{-5}$ molar mixture of terrylene/p-terphenyl in toluene. The solution is heated until both compounds are dissolved and then isopropanol is added as a reprecipitation agent. This procedure results in terrylene doped p-terphenyl crystals of platelet morphology as seen in Fig. 2(a), which shows a scanning electron microscope (SEM) image of such crystals deposited on a silicon substrate. The majority of crystals have base dimensions in the range of 200 to 2000 nm and a base width to height ratio of 2.5:1 to 5:1 as determined by atomic force microscopy (AFM) measurements. The single-crystalline nature of these crystals has been verified by performing selected area electron diffraction (SAED) measurements using a transmission electron microscope (TEM), see Fig. 2(b). SAED also indicates that the substrate-supported crystal platelet base face is of (001) orientation, i.e. the crystal’s c-axis is perpendicular to the substrate. It is known that the dipole moment of the transition to the lowest electronically excited state in terrylene is linear and lies along the long axis of the molecule \[ [53] \]. When inserted into a p-terphenyl crystal, the molecule’s long axis and thus the dipole moment lie nearly parallel to the crystal’s c-axis and therefore in our configuration nearly perpendicular to the substrate.

A terrylene-doped p-terphenyl nanocrystal is deposited on the nanofiber by a drop-touch method: a drop of the suspension of doped p-terphenyl nanocrystals is briefly brought into contact with the nanofiber via a pipette. During this process, the transmission of the excitation laser and the fluorescence of the nanofiber is monitored with a power meter and a spectrometer, respectively. When a doped crystal has adhered to the nanofiber surface during the contact with the suspension droplet, a typical fluorescence signal and some loss
in transmission is observed. As crystals of a variety of sizes are produced during our growth process, we have to post-select the size of the deposited crystal. However, since the largest crystals sediment faster, we usually find suitably small ones in the suspension supernatant. If it is nevertheless found that the transmission deteriorates too much during the deposition process, the crystal can be washed off with acetone and another one deposited.

The TOF is mounted on a steel holder with two NdFeB magnets. This ensures that the fiber is firmly held and that it stays intact during the cooling process down to cryogenic temperatures. This fiber set-up is mounted in the cold-pot of a custom-made cryostat that can cool the sample to 4 K. To ensure efficient thermalisation of the nanofiber and the crystal with the walls of the cold-pot, helium buffer gas at a pressure of a few mbar is introduced into the cold-pot before cool-down, after it has been evacuated.

To excite the molecules, light of a dye laser (Spectra-Physics Matisse-DS) is coupled into the optical fiber that connects to the TOF and that enters the cryostat via a teflon feed-through [Fig. 1(a)]. To compensate for intensity fluctuations and drifts, the laser beam is sent through an acousto-optic modulator (AOM) and partly onto a photodiode (PD) and is actively intensity stabilised. A fraction of the Stokes-shifted laser induced fluorescence (LIF) of the molecule is collected by the nanofiber, fiber-guided out of the cryostat, and can be monitored by a spectrometer (Shamrock SR-303i, Andor Technology) or single photon counting modules (SPCMs) at either end of the optical fiber. Contributions from the excitation light and fluorescence on the 00ZPL are filtered out by a long-pass (LP) filter. Together with a short-pass (SP) filter to block Raman scattering from the fiber, this leaves a transmission window in the range of 630–650 nm. Due to the inhomogeneous line shifts induced by the host crystal matrix, single molecules can be spectrally selected with the narrowband dye laser if the terrylene concentration in the host crystal is small enough. Figure 3 shows the fluorescence excitation spectrum of a molecular ensemble in the X4 orientation. The excitation line of what has been verified to be a single molecule is highlighted in red.

To characterize the optical interface created by individual molecules and the optical nanofiber, we have investigated several molecules that are located in the same nanocrystal. The transition frequencies of solid state quantum emitters are known to be very sensitive to the environment, which can be a favourable effect if con-
trolled well \cite{60} or lead to unwanted spectral diffusion. Therefore, the stability of individual molecular spectra is recorded by consecutively scanning the same spectral line and monitoring the center frequency over time. An example of this can be seen in figure 4. This shows the stability of a molecular spectrum over the timescale of minutes. This stability is beneficial in comparison to other solid state emitters, which may require active stabilisation of the resonance frequency \cite{61, 62}. By cooling the doped crystal to 2.4 K, we can also readily achieve lifetime-limited spectra of our single molecules recorded on either end of the TOF, as is experimentally shown in the supplemental material.

\[ g^{(2)}(\tau) = A \left( 1 - \left[ \cos(\nu\tau) + \frac{3\Gamma}{4\nu} \sin(\nu\tau) \right] \exp\left( -\frac{3\Gamma\tau}{4} \right) \right) \]

with \[ \nu = \sqrt{\Omega^2 - \left( \frac{\Gamma}{4} \right)^2} \]

An incoherent Poissonian background \( B \) added to a signal with average intensity \( \langle I \rangle \) reduces the contrast of the intensity correlation measurements. This is taken into account when analyzing our data with the modified intensity correlation function \cite{63}

\[ g^{(2)}_B(\tau) = 1 + \frac{\langle I \rangle^2}{(\langle I \rangle + B)^2} (g^2(\tau) - 1). \]

We fit eqn. (3) to the data for different excitation powers while using \( \Gamma \) as a global fit parameter for all measurements on a given molecule. This yields the respective Rabi frequencies and the non–power broadenend homogeneous linewidth \( \Gamma \) of the molecule. From these fits we also obtain the saturation intensity \( I_S \) of the molecule as \( I/I_S = 2\Omega^2/\Gamma^2 \). Figure 6 shows this expected linear increase of the squared Rabi frequency as a function of excitation power for three different molecules in the same crystal. The error bars obtained from fitting the intensity correlation measurements are smaller than the depicted datapoints in Fig. 6. From the fits, the saturation power \( P_S \) corresponding to \( I_S \) is obtained. We obtain saturation powers for molecule A of \( P_S = 1.8 \pm 0.3 \) nW, molecule B of \( P_S = 4.3 \pm 0.8 \) nW and molecule C of \( P_S = 0.5 \pm 0.1 \) nW. The biggest contribution to the error in the saturation powers arises from the uncertainty in determining the power in the fiber. We convert \( P_S \) to the maximum intensity at the surface of the nanofiber by considering the fundamental quasi-linearly polarised HE\(_{11}\) mode that is supported by our optical nanofiber with a diameter of 320 nm \cite{38}. Without the
Figure 5. Fluorescence intensity correlation measurements of molecule C for different excitation powers (from bottom to top: 0.7 nW, 2.9 nW, 7.4 nW, 11.1 nW). The coincidences were recorded for 1000 s and the time resolution for the data is 1 ns. With increasing excitation power, the onset of Rabi oscillations and an increase in incoherent background scatter is clearly seen. The inset shows the $g^{(2)}$-measurement at 2.9 nW normalised to the steady-state correlations of the fluorescence of the molecule, clearly indicating a single emitter.

exact knowledge of the orientation of the molecule’s transition dipole moment and its distance from the nanofiber surface, this gives an upper limit for the saturation intensity. The measured saturation intensities are $I_S < 1.8 \text{ Wcm}^{-2}$ for molecule A, $I_S < 4.3 \text{ Wcm}^{-2}$ for molecule B and $I_S < 0.5 \text{ Wcm}^{-2}$ for molecule C. These results compare well with results obtained by other groups who studied tereylene in bulk p–terphenyl [42, 64]. To our knowledge a saturation intensity of $I_S < 0.5 \text{ Wcm}^{-2}$ as for molecule C is the lowest measured so far for tereylene in p–terphenyl. As opposed to measurements on tereylene in bulk p–terphenyl using a confocal microscope, the excitation light in our case enters through the side of the thin host crystal platelets. As the transition dipole moment of the molecules lies nearly perpendicular to the base of these platelets, this suggests an improved overlap between the polarization of the nanofiber–guided excitation light and the transition dipole moment of the molecules. An independent measurement of the saturation power is obtained by recording the resonant fluorescence rate $R_{\text{LIF}}$ as a function of excitation power as plotted in Fig. 7, which includes a fit to $R_{\text{LIF}} = R_{\infty} \frac{P}{1+P/P_S}$. The error bars on the fluorescence rate that are obtained by taking the standard error of the amplitude from fits over several molecular spectra are smaller than the depicted datapoints. This measurement yields a saturation power of $4.8 \pm 1.4 \text{ nW}$ for molecule B, where the error stems from the fit and from the uncertainty in the excitation power inside the fiber. This translates into a saturation intensity of $I_S < 4.8 \text{ Wcm}^{-2}$ for molecule B in good agreement with $I_S < 4.3 \text{ Wcm}^{-2}$ as obtained with the HBT setup. We performed a further measurement on a fourth molecule (molecule D) that yielded a lower fluo-

Figure 6. Squared Rabi frequency as a function of excitation power for molecule A (red, squares), B (blue, circles) and C (black, triangles) and corresponding linear fits.

Figure 7. Saturation of the resonant fluorescence intensity $R_{\text{LIF}}$ as the excitation laser power is increased for molecules B (blue, circles) and D (green, diamonds) and corresponding fits.
The nanofiber interface is then given as $\eta_{\text{eff}}$ for fluorescence excitation and detection via the nanofiber interface. The molecules are also detected by coupling to the nanofiber–guided modes as efficiently. Alternatively, the alignment between polarization of the excitation light and the dipole moment of molecule D may be less favorable. Although all molecules are embedded in a single crystal and therefore have the same orientation, the inherent birefringence of the host crystal can cause a less favorable alignment. The phase shift between the corresponding polarization components is $\Delta \phi = 2\pi \Delta n L / \lambda$, where $\lambda$ is the vacuum wavelength, $L$ the propagation distance and $\Delta n$ the effective birefringence that can reach 0.32 in our case. The efficiency of exciting the different molecules is given by $\eta_{\text{abs}} = \sigma / A_{\text{eff}}(x,y)$, where $A_{\text{eff}}(x,y)$ is the effective mode area at the position of the molecule. More details can be found in the supplementary information. The molecules are also detected via the nanofiber interface and hence the overall efficiency for fluorescence excitation and detection via the nanofiber interface is then given as $\eta_{\text{LIF}} = \eta_{\text{abs}} \beta$, where $\beta = \Gamma_g / \Gamma_{\text{sc}}$ is the coupling efficiency of dipole radiation to the nanofiber modes. This coupling efficiency depends on the radiated wavelength, distance and orientation of the dipole with respect to the nanofiber surface. Here, $\Gamma_g$ is the scattering rate into guided modes and $\Gamma_{\text{sc}}$ is the total scattering rate of the dipole. For our case of a nanofiber with 160 nm radius and Stokes shifted fluorescence in the range of 630–650 nm, we calculated this coupling efficiency for a radially, azimuthally and axially oriented dipole following [38], see Fig. 8.

Since the power needed to saturate molecule C is the lowest yet measured for terrylene in p–terphenyl, we assume that this molecule is very close to the surface of the nanofiber. An upper limit for the other molecules from the nanofiber surface can then be estimated by comparing their saturation intensities. Because the host crystal is birefringent, we only give an upper limit on the radial distance between the different molecules. Figure 9 shows the calculated efficiency of fluorescence excitation via the nanofiber interface for different positions of the dipole with respect to the nanofiber surface. The positions are chosen to lie within the volume of a platelet crystal with its base on the nanofiber surface. The molecular dipole is oriented perpendicular to the crystal’s base and excited by quasilinearly polarised light via the nanofiber–based interface. Assuming unpolarised light instead of quasilinearly polarised light affects the relative distances between the different molecules by less than 3%. We did not incorporate the refractive indices of the crystal into this model because they would make the local efficiencies very dependent on the crystal’s geometry and we are only interested in assigning the maximum radial distance of the measured molecules. The maximum radial distances of the investigated molecules are depicted in their respective colours by dashed contours [Fig. 9]. These results show that they radially all lie within less than 481 nm of each other and less than 291 nm from the nanofiber surface. This translates into coupling efficiencies for a radial dipole to the nanofiber mode between 5–30% [Fig. 8]. This means that our set-up can be a superior choice for coupling single photons to single mode optical fibers compared to using conventional confocal microscopes [65, 67] and thus opens the way for fully fiber–coupled single photon sources. It is also an important step towards strong coupling of single molecules to optical waveguide structures.

Summarizing, we have shown how single molecules can be optically interfaced via the evanescent field surrounding an optical nanofiber. This is an important addition to the toolbox of quantum emitters such as atoms [34, 68, 69], quantum dots [33, 65] and NV centers in nanodiamonds [41, 70] that have been fiber-integrated by coupling to optical nanofibers. Each of these systems has its own intrinsic advantages for their usage in quantum networks. Single molecules in solids are efficient quantum emitters that come in a large variety of emission wavelengths. This makes them suitable to be interfaced with other quantum emitters [1]. They have an advantageous level structure for the implementation of triggered single photon sources [43, 71] and have proven their versatility in quantum optics [72, 74]. Further, single waveguide–coupled molecules allow the investigation of photon–mediated interactions between two quantum emitters even when they are separated by much more than the excitation wavelength [75, 77]. These interactions can be further enhanced by using a nanofiber between two fiber Bragg gratings and thereby realizing a high–Q cavity [78]. Single molecules that are coupled to the evanescent field of optical nanofibers therefore not only offer a rich experimental platform for investigating
entanglement and correlations between quantum emitters, they also provide a means for implementing components of quantum networks such as fiber-coupled single photon sources [32, 79, 80] or photon sorters [81, 82].

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Figure 9. The excitation efficiency \( \eta \) for a molecular dipole at different positions with respect to the nanofiber. The spatial coordinates are chosen assuming a platelet crystal that is lying with its base on the nanofiber surface. The contact point of nanofiber and crystal is at the origin of this reference frame. The efficiency is normalised to the maximum excitation efficiency on the nanofiber surface of 13% (see supplementary information). The maximum radial distances for the different molecules are indicated by dashed colored lines (A = red, B = blue, D = green, from bottom to top). The reference molecule C is shown as a black dot and is assumed to be located directly on the nanofiber surface at position \( (x, y) = (0, 0) \).

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