Electrical Control of Lifetime-Limited Quantum Emitters Using 2D Materials

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Supporting Information

ABSTRACT: Solid-state quantum emitters are a mainstay of quantum nanophotonics as integrated single-photon sources (SPS) and optical nanoprobes. Integrating such emitters with active nanophotonic elements is desirable in order to attain efficient control of their optical properties, but it typically degrades the photostability of the emitter itself. Here, we demonstrate a tunable hybrid device that integrates state of the art lifetime-limited single emitters (line width ∼40 MHz) and 2D materials at subwavelength separation without degradation of the emission properties. Our device’s nanoscale dimensions enable ultrabroadband tuning (tuning range >400 GHz) and fast modulation (frequency ∼100 MHz) of the emission energy, which renders it an integrated, ultracompact tunable SPS. Conversely, this offers a novel approach to optical sensing of 2D material properties using a single emitter as a nanoprobe.

KEYWORDS: Single molecules, 2D materials, electrical control, single photon source, Stark effect, lifetime-limited line width

Hybrid nanophotonic systems blend the strengths of distinct photonic elements to strongly enhance light–matter interactions in integrated photonic circuits. In these systems, narrow-line width quantum light emitters play a key role as single-photon sources (SPS) which interact with their nanoscale environment. Controlling these interactions provides versatile SPS tuning required for coupling quantum resources. Integrating nanoscale light emitters with two-dimensional (2D) materials is motivated by the rich physics of near-field interactions and new hybrid light-matter states. This approach unites integrated solid-state SPS such as nitrogen vacancy centers, quantum dots, and single molecules with the diverse optoelectronic properties of 2D materials that facilitate emitting, controlling and detecting light at the nanoscale. In such hybrid devices, quantum emitters can be integrated at subwavelength separation to the 2D interface to achieve efficient near-field coupling, which modifies the emitter’s radiative decay rate or transition energy. Recent experimental studies integrated 2D materials with ensembles of broadband emitters to demonstrate electrical and electromechanical tuning of the decay rate by controlling nonradiative energy transfer (nRET) or the energy flow to confined electromagnetic modes such as 2D polaritons. Therefore, hybrids of 2D materials and SPS have the potential for in situ control of the conversion and channelling of single photons at the nanoscale. So far, these studies have been limited to ensembles and broad line width emitters. Integrating bright and narrow quantum emitters in such systems paves the way toward a tunable quantum light-matter interface, which is an essential ingredient for integrated quantum networks.

Here, we demonstrate hybrid integration of 2D materials (semitetallactic graphene or semiconducting MoS2) with single, lifetime-limited quantum emitters in nanocrystals to provide active emission control. Using the 2D materials as transparent electrodes, we show broadband Stark tuning of the emission energy over 40 000 times the emitter line width and fast modulation of the emitter’s optical resonance on the time scale of 20-200 μs. The device provides versatility allowing tuning over more than 500 GHz in the emission energy and fast modulation up to 1 GHz.

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of its radiative lifetime. Such tuning can mitigate inhomogeneous broadening in solid-state environments to enable resonant and synchronized interaction between distinct quantum systems and allows for controlled coupling of narrowband quantum emitters to broadband nanophotonic circuitry. Our approach is particularly suited for making integrated devices: although the 2D material is just tens of nanometers from the quantum emitter, we observe only weak emission line width broadening and spectral diffusion. In contrast, emitters close to bulk transparent electrodes such as ultrathin metal films suffer strong emission quenching, which hampers nanoscale integration. Furthermore, we find that the deposition process required to integrate a transparent conducting oxide inhibits molecular fluorescence, which highlights the potential of 2D materials for integration with ultrasensitive quantum emitters. At the same time, we show that a single quantum emitter can be used as a transducer of the 2D materials’ electronic properties.

We chose single dibenzoterrylene (DBT) molecules as bright, photostable single-photon sources emitting at 785 nm (1.58 eV) with lifetime-limited line width (~40 MHz) at 3 K even when hosted in a submicron environment. Experimentally, we perform scanning laser spectroscopy to address individual DBT molecules at subwavelength separation to a 2D metallic or semiconducting interface. The device comprises DBT molecules embedded in anthracene nanocrystals (see Supporting Information, section S1), interspersed in a thin poly(vinyl alcohol) (PVA) film on a Si++/SiO2 substrate (Figure 1a). MoS2 or graphene flakes are placed on top of the polymer film by a dry transfer technique and electrically contacted by gold electrodes. This transfer process is performed at room temperature and without other nanofabrication steps, which could potentially degrade emission. Spatial maps of DBT ensemble fluorescence (see Methods) at 3 K show bright, localized emission beneath both 2D materials (Figure 1b).

A simplified DBT energy level scheme is shown in the inset of Figure 1a. The transition of interest is the zero-phonon line (ZPL) between the ground vibrational levels of the electronic ground and excited states. Upon resonant excitation of this transition, the molecule relaxes either to the electronic ground state (30–40% of emission) or to a higher vibrational state, thereby emitting a red-shifted photon. Scanning laser spectroscopy (see Methods) on a pristine nanocrystal (Figure 1c) at 3 K reveals a series of sharp peaks from a DBT ensemble. This peak dispersion arises from local variations of strain and charge and enables spectrally addressing a single molecule. The narrowest peaks in such ensembles display a typical line width of $\Gamma/2\pi = 43 \pm 7$ MHz.

Figure 1. Single molecules integrated with 2D materials. (a) Hybrid device schematic. Atomically thin layers of graphene and MoS2 cover fluorescent molecules embedded in a PVA film (h_{PVA} = 300 nm) on SiO2 (h_{SiO2} = 285 nm). Single molecules are resonantly excited (inset) and their red-shifted fluorescence detected with a single-photon counting module (SPCM). Electric fields are controlled by applying DC ($V_g$) and AC ($\delta V_g$) potentials to the Si++ backgate. (b) Top to bottom: optical micrograph, AFM topography, and DBT emission map for MoS2 (left column) and bilayer graphene (right column) devices. White dashed lines outline the flakes. Scale bars are 20 μm. (c) Top panel: fluorescence excitation spectrum of an ensemble of single molecules in an uncovered nanocrystal at 3 K. Bottom panel: detail of two emission peaks with Lorentzian line shape (solid line), the narrower peak shows a fwhm of $43 \pm 7$ MHz. (d) Antibunching measurement for resonant excitation of a single peak as shown in part c. The solid line is a fit to the data using a second-order correlation function.
To confirm that we can address single molecules in a nanocryystal, we measure statistics of photon emission in Hanbury-Brown and Twiss (HBT) configuration (Figure 1d) and fit the normalized data with the second-order intensity autocorrelation function\(^{10}\) \(g^2(\tau) = 1 - C e^{-\tau / \Gamma_{\text{obs}}}\), where \(C\) is the contrast. At zero delay time, we find \(g^2(0) = 0.04 \pm 0.02 < 0.5\), which is signature of a single-photon source. We extract an excited state lifetime\(^{16}\) of \(\tau_{\text{DBT}} = 4.7 \pm 0.5\) ns, implying a lifetime-limited line width \(\Gamma_{\text{D}} / 2\pi = 34 \pm 5\) MHz, comparable to values observed in bulk anthracene crystals.\(^{37}\)

Therefore, single molecules in our device can exhibit lifetime-limited line width within the measurement error. In a solid state environment, this line width can be broadened by dephasing and near-field interactions, which can hamper the performance of emitters in nanostructured devices.\(^{36,40}\)

In our device, a 2D material is placed in the near-field of a single emitter (separation \(d \ll \lambda\)). In this regime, near-field interactions such as nonradiative energy transfer (nRET) from the molecule to the 2D material\(^{31}\) and Casimir–Polder (CP) energy level shifts\(^{8}\) are expected to occur. Both interactions scale divergently with separation as \(d^{-3}\), in contrast to \(d^{-1}\) for bulk interfaces.\(^{12}\) The nRET process leads to a line width broadening, while the CP energy level shift is a quantum effect related to the modification of vacuum fluctuations by the 2D interface. At fixed emitter–2D material separations, we can quantify the nRET contribution due to the presence of the 2D material by its impact on the emission line width. The CP contributions are difficult to quantify because for one specific emitter, the emission energy with and without the 2D material cannot be measured independently.

By measuring the emission line widths of over 500 molecules (Figure 2) in three different configurations (uncovered, covered by bilayer graphene (BLG)/MoS\(_2\)) in different samples, we quantify the effect of proximity to 2D materials on emission line widths. We quantify the most probable line width \(\Gamma\) in each case by fitting the line width histogram with a Smirnov distribution,\(^{12}\) which describes single molecule line widths distributions perturbed by long-range coupling to two-level fluctuators, e.g., in polymers.\(^{43,44}\) For uncovered nanocrystals, we obtain \(\Gamma_{\text{uncov}} / 2\pi = 62\) MHz, which is comparable to \(\Gamma_{\text{D}}\). This implies that the anthracene nanocrystal is a highly stable and crystalline environment,\(^{35}\) and most molecules within it do not experience significant spectral diffusion due to defects or proximity to the surface.\(^{35}\) While emitters covered by MoS\(_2\) experience extremely weak spectral broadening (\(\Gamma_{\text{MoS}} / 2\pi = 65\) MHz), they are significantly broadened when covered by BLG (\(\Gamma_{\text{BLG}} / 2\pi = 108\) MHz) and their line width distributed over a larger range. We attribute this broadening to higher nRET efficiency to gapless graphene as electronic transitions can be optically excited over a large range of energies and in particular at \(\hbar\omega_{\text{ZPL}}\). In contrast, MoS\(_2\) has bandgap and excitonic resonances at energies > \(\hbar\omega_{\text{ZPL}}\), resulting in weak nRET, which preserves the narrow line width. These results show that neither the presence nor the integration process of 2D materials have a strong detrimental impact on the emission properties. In contrast, we find that the deposition of a commonly used transparent electrode material such as ITO results in the complete disappearance of DBT emission (see Supporting Information, section S2), most likely due to thermal damage to the nanocrystal during deposition. This highlights the weakly invasive nature of 2D material integration, which makes it particularly suitable for fragile quantum emitters.

To confirm that emission stability in time is preserved after 2D material integration, we measure DBT spectra over time for the three configurations (uncovered, covered by graphene/MoS\(_2\)) as shown in Figure 2b. In each case, we observe stable emission intensity and fluctuations of ZPL frequency and line width below 42 MHz (see Supporting Information, section S3). Slightly higher fluctuations of \(\omega_{\text{ZPL}}\) — possibly induced by charge fluctuations — are resolved in the case of MoS\(_2\). Overall, these results show that 2D semiconductors are particularly suitable for integration with sensitive quantum emitters at nanoscale proximity, introducing negligible perturbation in the photostability.

We now turn to electrical manipulation of single emitters. To achieve Stark\(^{45}\) tuning of DBT emission energy, we apply an electric potential \(V_g\) over the capacitor formed by the 2D electrode and the Si\(^{++}\) back-gate, separated by a PVA/SiO\(_2\) layer (see Figure 1a). We reach comparatively large\(^{46,47}\) field strengths above 2 MV cm\(^{-1}\) before dielectric breakdown takes place. DBT ensemble spectra below BLG at different \(V_g\) (Figure 3a) show a large, dominantly quadratic shift of the emitters in the whole ensemble on the order of hundreds of GHz (\(\sim 10^4 \Gamma_0 / 2\pi\)), comparable to the inhomogeneous broadening of the entire ensemble.\(^{35}\) While this tuning range is comparable to state-of-the-art devices employing semiconductor quantum dots\(^{47}\) and diamond defects,\(^{48,49}\) we highlight that our device displays emission line widths within a smaller footprint, which is attractive for nanophotonic integration. We also verify that DBT emission energy remains stable under large applied electric fields by repeating stability measurements as shown in Figure 2b up to large values of \(V_g\) (see Supporting Information, section S3). We find small, gate-dependent emission energy drifts (<3 MHz/s), likely due to small leakage currents in the device, which can be reduced by using a smaller 2D electrode area or by active feedback.\(^{50}\)

For a centrosymmetric molecule such as DBT, we expect a quadratic Stark shift with linear contributions arising from

![Figure 2. Emission broadening and time stability in the presence of 2D materials. (a) Line width distribution for uncovered DBT (green), DBT covered by monolayer MoS\(_2\) (red), and bilayer graphene (gray). Solid lines are fits to the data as described in the main text. (b) Time trace of single DBT molecule emission (3 s/line) for uncovered DBT (bottom) and DBT covered by monolayer MoS\(_2\) (middle) and bilayer graphene (top).]
dipole moments induced by distortion of the molecule’s insertion site. The Stark shift is modeled as
\[ \Delta \omega_{\text{ZPL}} = a|\vec{E}| + b|\vec{E}|^2, \]
where \( a \) and \( b \) are the linear and quadratic Stark coefficients, and functions of the dipole moment and polarizability change, respectively, between ground and excited state. Here, \( \vec{E} = L\vec{E}_{\text{ext}} - \vec{E}_0 \) is the net local electric field experienced by the molecule upon application of an external field \( |\vec{E}_{\text{ext}}| = V_g/h_{\text{PL}} + h_{\text{SiO}_2} \sim 600 \text{ nm} \) with a
correction \( \tilde{E}_g \) that accounts for intrinsic electric fields, e.g., due to trapped charges and work function differences.\(^{50}\) We also include a local field correction factor \( L = \frac{\varepsilon + 2}{3} = 1.7 \), derived from anthracene’s averaged isotropic permittivity\(^{31}\) \( \varepsilon = 3.1 \). Fitting detuning data for different emitters, we extract typical Stark coefficients of \( a \sim 300 \text{ MHz} / (\text{cm}^{-1}) \) and \( b \sim -0.15 \text{ MHz} / (\text{cm}^{-1})^2 \) (see Supporting Information, section S4), comparable to reported results.\(^{34, 35}\) The observed quadratic Stark shift shows that \( E_{\text{BL}} \) is linear in \( V_g \), as is expected for a metallic electrode such as BLG (Figure 3b).

In contrast, emitters under MoS\(_2\) deviate from the parabolic detuning below \( V_{g,0} \sim -30 \text{ V} \) (Figure 3b), where the detuning flattens and \( \Delta \omega_{\text{ZPL}} \) is almost independent of \( V_g \). We attribute this deviation to a gate-induced change of resistivity:\(^{52}\) as the charge carrier density in a semiconductor changes more abruptly with \( V_g \), we expect a sublinear dependence of \( E_{\text{ind}}(V_g) \) as excess charge carriers are depleted in MoS\(_2\). Electrostatic doping of the MoS\(_2\) electrode is independently confirmed by gate-dependent photoluminescence (Figure 3c,d). The ratio of charged (X) to neutral (X') exciton PL intensities increases with \( V_g \) due to n-doping of the MoS\(_2\) electrode. We also observe a broad emission peak at lower energy for negative \( V_g \)—associated with emission from defects in MoS\(_2\)—which overlaps with \( h \omega_{\text{ZPL}}. \) With the appearance of this defect band, we observe a line width broadening \( f_{\text{DBT}} \) below \( V_{g,0} \) for some emitters, which is absent in the case of a metallic BLG electrode (Figure 3e). This suggests that defects could act as n-RET acceptors, leading to emission line width broadening of emitters close to MoS\(_2\).

Our system combines large Stark tuning with lifetime-limited emitters that potentially enable adiabatic control up to a frequency set by the emitter’s natural line width \( \Gamma_f \). We investigate the range of this dynamical control by applying oscillating potentials \( V_{\text{tot}} = V_g + \delta V_g(f_{\text{AC}}) \) to graphene and MoS\(_2\) electrodes. For fast modulation frequencies compared to the spectral acquisition time \( (f_{\text{AC}} \gg f_{\text{meas}}^{-1} \sim 0.1 \text{ Hz}), \) DBT emission peaks show a splitting \( \sigma(f_{\text{AC}}) \) (Figure 4a) proportional to the modulation amplitude \( \delta V_g \) and the local Stark slope \( \delta \omega_{\text{ZPL}} / \delta V_g \):

\[
\sigma = \frac{\partial \omega_{\text{ZPL}}}{\partial V_g} \delta V_g \Gamma_f
\]

Here, \( T(f_{\text{AC}}) \) is the transmission of the RC-low-pass filter governed by the effective device resistance \( R \) which includes contact resistance as well as the sheet resistance of the 2D material and backgate capacitance \( C \) (see Supporting Information, section S5). At fixed \( V_g \) and \( f_{\text{AC}} \), the DBT emission spectra reflect the oscillation turning points of the modulating waveform (Figure 4a). Under square modulation with a fixed amplitude \( \delta V_g \), the splitting vanishes with increasing \( f_{\text{AC}} \) for both MoS\(_2\) and graphene electrodes (Figure 4b), albeit at a lower frequency for MoS\(_2\). From this measurement, we extract \( T(f_{\text{AC}}) \) (eq 1) and confirm low-pass behavior with a characteristic cutoff frequency \( f_{\text{low}} \) (Figure 4c). Interestingly, \( T(f_{\text{AC}}) \) shows a strong gate voltage \( V_g \) dependence for MoS\(_2\) electrode, which is not observed for graphene. This dependence is quantified by \( f_{\text{low}} \) which increases with \( V_g \) and saturates at \( \sim 5 \text{ kHz} \) \( \ll \Gamma_f \) for \( V_g > 0 \text{ V} \) (Figure 4d). We attribute this behavior to a change of sheet resistivity and contact resistance due to gate-induced electrostatic n-doping\(^{53}\) of the MoS\(_2\), consistent with PL measurements (Figure 3c,d). As a result, \( f_{\text{low}} \propto (\text{RC})^{-1} \) increases as we sweep \( V_g \) from negative to positive values.

To capture the gating efficiency of our 2D electrodes, we extract transmission \( T \) at fixed \( f_{\text{AC}} = 10 \text{ kHz} \) varying \( V_g \) (Figure 4e, and Supporting Information). The strong reduction of \( T \) for \( V_g < 0 \text{ V} \) is consistent with a large gate-induced change of resistance in the semiconducting MoS\(_2\) device, while graphene’s metallicity maintains \( T \approx 1 \) over the full \( V_g \) range. Therefore, graphene enables Stark tuning of emitters over a large energy range and at high frequency at the expense of weak line width broadening. To extend this modulation bandwidth, we reduce the gate capacitance \( C \) by using a thicker PVA dielectric \((h_{\text{PVA}} = 800 \text{ nm})\). Then, we modulate \( h \omega_{\text{ZPL}} \) around a fixed excitation energy by applying \( \delta V_g \) at \( f_{\text{AC}} = 100 \text{ MHz} \sim \Gamma_f / 2\pi \) to a graphene electrode. Using a time-correlated single-photon counter synchronized to the modulation \( \delta V_g \) we observe a periodic oscillation of emission intensity (Figure 4f) with a modulation depth of \( \sim 50\% \) (measurement time 30 s). This oscillation is a signature of the excitation laser periodically exploring a fraction of the emitter’s absorption line. These measurements show that the dynamical modulation bandwidth of our devices, determined by the 2D electrode material and device geometry, approaches \( \Gamma_f \). Conversely, single emitters act as local nanoprobes of the 2D material’s electronic properties.

In conclusion, we demonstrate a hybrid device where 2D materials are integrated with lifetime-limited single-photon emitters to achieve broadband and fast emission energy tuning. Our results highlight the potential of atomically thin electrodes for integration with sensitive quantum emitters in a nanoscale device without perturbing narrow emission line width, in contrast to commonly employed bulk transparent electrodes. At high frequencies, Stark modulation reveals low-pass transmission behavior related to the 2D materials’ sheet resistivity. Using a graphene electrode, we show emission energy modulation at frequencies approaching the emitters’ line width. Our device thus provides resonant tuning and high-frequency modulation of SPS on chip, required for obtaining indistinguishable and synchronized single photons. Finally, our device is a platform for studying novel forms of light-matter interaction such as plasmon-polaritons in graphene\(^{55}\) and exciton-polaritons in TMDs\(^{10}\) at the single excitation level. Conversely, strong coupling to such excitations could allow the observation of normally forbidden higher order transitions of the emitter.\(^{54}\)

**Methods. Device Fabrication.** We make a suspension of anthracene nanocrystals hosting DBT molecules in PVA by reprecipitation. The suspension is spin-cast onto a p-doped Si wafer coated with 285 nm thermally grown SiO\(_2\). Electrodes are deposited onto the coated chip by thermal evaporation of 100 nm Au through a shadow mask. Nanocrystals close to the electrode displaying DBT fluorescence are localized at room temperature using off-resonant wide-field illumination. Bulk MoS\(_2\) and graphite are exfoliated mechanically using commercial polydimethylsiloxane (PDMS) sheets. Mono- and bilayer flakes of both materials are identified optically by absorption contrast (see Supporting Information, section S6) and transferred onto anthracene nanocrystals in PVA by dry stamp transfer.

**Electrical Device Actuation and Optical Readout.** Measurements are performed under vacuum in a cryostat at 3 K. Our hybrid devices are actuated electrically using a low-noise voltage source and an arbitrary waveform generator to
provide DC and AC voltages. Using a custom-built confocal microscope to locally illuminate the device, we excite single molecules with a tunable 785 nm laser with circular polarization at 5 nW, well below the saturation power of ∼20 nW (see Supporting Information, section S7). Red-shifted single photon emission is detected with a single-photon counting module (SPCM) combined with spectral ZPL filtering by a long-pass filter. Emission maps are made using pseudobroadband excitation of DTB ensemble fluorescence by fast modulation (200 Hz) of the laser detuning compared to the SPCM integration time (∼10 ms). MoS2 photoluminescence is excited using a 532 nm laser and detected with a spectrometer (10 s integration time).

**ASSOCIATED CONTENT**

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Nanocrystal characterization, impact of ITO deposition on molecular emission, emission stability, Stark coefficients, electrical low-pass model of the device, 2D material identification, and emission saturation behavior (PDF)

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**Author Contributions**

K.G.S., A.R.-P., C.T., and F.H.L.K. conceived the experiment. K.G.S. and C.C. made the samples with the support of S.P., P.L., and C.T. Measurements and analysis were performed by K.G.S., C.C., and A.R.-P. K.G.S, A.R.-P. and F.H.L.K. wrote the manuscript with critical comments from all authors.

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**Notes**

The authors declare no competing financial interest.

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