QSS45 - Maja Colautti, Tom Darras, Daniel Goncalves Romeu - Questions & Answers

Maja Colautti, Tom Darras, Daniel Goncalves Romeu

Table of Contents

1. Maja Colautti

Bill Phillips: Concerning brightness, I would have expected a molecule to pump into a “dark” state after a few emissions. Why doesn’t it?

MAJA: In general, organic dye molecules do exhibit strong blinking, which dramatically decreases fluorescence. The crucial aspect of achieving a photostable molecular emitter is choosing a suitable combination of guest flourophore and host matrix. In particular, for dibenzoterrylene (DBT) molecules in anthracene (Ac) crystals, the first singlet excited state of the fluorophore is lower than both the singlet and the triplet state of the host consequently reducing the inter system crossing (ISC) probability, which is directly connected to the occurrence of dark states. Specifically, measured values of ISC yield for this system are on the order of 10^{-7} (Nicolet et al, ChemPhysChem 8 (2007)), the smallest ever reported for organic molecules, with a very short triplet lifetime on the order of 10us. Such photophysical properties make DBT in Ac an especially suitable system for single molecule spectroscopy and bright single-photon emission. This is attested also by the high detected counts reported in several articles (e.g. Lombardi et al, Adv. Quantum Technol. (2019); Colautti et al, Adv. Quantum Technol. (2020))

Do molecules bleach? how long can you work with the same molecule?

MAJA: Our molecules do not bleach, and the explanation why is similar to the previous answer: while in general organic dye molecules bleach after few fluorescence cycles, once the fluorophore is protected from photo-oxydation by a suitable matrix, photochemistry is strongly suppressed. At ambient conditions the Ac matrix undergoes sublimation, however DBT in Ac might not photobleach even after 10 hours of constant excitation (Toninelli et al, Optics Express 7 (2010)). We typically prevent from Ac sublimation by covering the sample with a thin polymer film of polyvinyl alcohol, for example. Anyway, when the system is cooled at cryogenic temperatures Ac sublimation is totally suppressed hence ensuring ideally infinite protection, and indeed in this case we can typically work for weeks on the very same molecule.

Is there a single molecule that emits the light, or is this a bunch of them at the same time in these nanocrystals? How do you prepare that?

MAJA: We can control the average amount of molecules in the single nanocrystal. The fabrication of nanocrystals consists in the reprecipitation in an aqueous suspension, following the injection of a mixed Ac and DBT solution into sonicating water. In particular, the Ac and DBT solution is a mixture of DBT in toluene and of Ac in acetone. By controlling the portion of the two we can control the average DBT density, and we have calibrated the procedure for achieving on average

one molecule per nanocrystal (for details see Pazzagli et al, Acs Nano (2018).

Would it be interesting to have many molecules in the same Nanocrystal?

MAJA: Yes, this is definitely interesting, especially for studying cooperative effects and many body physics. From a very practical point of view, having several molecules in the same crystal increases the statistics and consequently the probability of finding a molecule with the desired properties, e.g. the brightness or the emission frequency in case of coupling to a resonant structure. In this case, the individual molecule would be spectrally selected via resonant excitation and by exploiting the inhomogeneous broadening.

Is it possible to grow / host the organic molecules in an environment where the emission properties improve (e.g. another nanocrystal)?

MAJA: Yes, an advantage of organic molecules is indeed their flexibility also in terms of chemical engineering. One of the big current challenges is indeed about finding a suitable fluorophore/matrix combination to achieve a bright single-photon emission in the telecom. Changing the host matrix can be key to the compatibility with photonic structures, indeed while Ac is convenient for its stability and solid state at room temperature conditions, e.g. DBT in para-dichlorobenzene, which is liquid at room temperature, can be suitable to effectively fill micro-fluidic channels and then be solidified into crystals upon cooling (Rattenbacher et al, New J. Phys. 21 (2019)). Another interesting example for quantum sensing applications, is that Stark effect is quadratic in DBT in Ac, while a large linear Stark shift has been achieved embedding DBT in 2,3-dibromonaphtalene (DBN) (Moradi et al ChemPhysChem 20 (2019)).

2. Tom Darras

Could you explain where the information is encoded in the continuous-variable case? Is it in the phase of the Schrödinger cat state? what determines the "digitalness" of the cat state? Is it just the amplitude of the source coherent state compared to its quantum noise?

TOM: In the continuous variables case, we use a specific CV encoding which is the coherent state superposition basis. In that case you can define the |cat+> state as a logical |0> and |cat-> as a logical |1>, and you can define a CV qubit as a coherent superposition of the two $c0\,|cat+\rangle + c1\,e^{i\varphi}\,|cat-\rangle$. The information is thus encoded in both the qubit weights c0 and the phase φ. For example, in the case of an evenly-weighted qubit (c0 = c1) you recover a coherent state $e^{i\varphi}\,|\alpha\rangle$. If you are interested in the CV cat qubits, you can have a look at these two references where such types of states can be experimentally created: Neergaard-Nielsen, et al., Optical continuous-variable qubit, PRL, 105, 053602 Le Jeannic, et al., Remote preparation of continuous-variable qubits using loss tolerant hybrid entanglement of light, Optica, 5, 1012.

Why not encode the quantum information in the polarization state of the light rather than the 0 and 1 Fock states?

TOM: It is indeed much more attracting to encode the DV qubit in the polarization basis which is much more resilient to losses as compared to the Fock basis where imperfect transmission translates into logical bit flips. However, the polarization encoding requires to double the experimental resource for the DV mode generation as compared to the Fock basis encoding. This is a route we are pursuing in our lab, where we recently built a second type-II OPO in that prospect. The generation of hybrid entanglement between optical dual-rail polarization qubit and a coherent state has been theoretically studied in this paper:

- Kwon, et al, Generation of hybrid entanglement between a single-photon polarization qubit and a coherent state, PRA, 91, 012340

And an experimental implementation of such states has been reported with post-selection in this paper:

- Sychev, et al., Entanglement and teleportation between polarization and wave-like encodings of an optical qubit, Nat. Com., 9, 3672

Clearly each of discrete and continuous variables have their own advantages and disadvantages. How can you make sure you avoid combining the disadvantages in the hybrid case? For example, in the hybrid case, how can we avoid the downside you mentioned that continuous variable encodings are sensitive to perturbations?

TOM: Indeed, the distinction between the advantages and disadvantages of the two encodings is true only for certain protocols and in certain conditions, and depending on the task at hand it is very subtle to find the experimental tools that are the best suited. For instance, our cat state generation requires the use of a heralding, which makes the generation of the non-Gaussian CV state probabilistic. Thus in the hybrid case we have to make sure that the advantages obtained by combining the two approaches exceed the disadvantage that can appear. But is certain context we observe the emergence of improved protocols. I give here a list of references where such scenarios can be observed:

- Morin, et al., Witnessing trustworthy single-photon entanglement with local homodyne measurements, PRL, 110, 130401
- Takeda, et al., Deterministic quantum teleportation of photonic quantum bits by a hybrid technique, Nature, 500, 315-318
- Lee, et al., Teleportation of nonclassical wavepackets of light, Science, 332, 6027
- Andersen, et al., High-fidelity teleportation of continuous-variable quantum states using delocalized single photons, PRL, 111, 050504.

To avoid the downside of the sensitivity of the CV states to perturbations, a strategy could be to transfer its state to a DV qubit by teleportation, which could be either transferred in optical fibers, or stored efficiently in quantum memories.

What is the advantage of doing homodyne detection in detecting single photon with respect to single photon detectors?

TOM: For our applications, homodyne detection is often a more efficient measurement technique as compared to single photon detectors. Indeed, the quantum state to be detected on the single photon detectors are usually prone to spatial and spectral filtering that induces transmission losses between the generation and the measurement. In our case, the transmission up to the SNSPD is about 50% while well designed homodyne detections can present detection efficiencies of 85%. In addition, quantum state tomography with homodyne detection enable a reconstruction of the full density matrix of the state (and correction for detection losses can be applied for the reconstruction) while the tomography of quantum states using single-photon detectors are limited to a few elements of the density matrix. Yet, in our experiment, the heralding by the single-photon detector is crucial as it defines the temporal mode of our state, and cannot be replaced by a homodyne detection so that purpose. Finally, and more subtly, homodyne detection can be used to perform quadrature conditioning which can mimic in specific condition a measurement of vacuum, which is hard to perform precisely with single-photon detectors. Details on the use of homodyne detection for improved protocols in our group can be found in the following references:

- Morin, et al., Witnessing trustworthy single-photon entanglement with local homodyne measurements, PRL, 110, 130401
- Le Jeannic, et al., Remote preparation of continuous-variable qubits using loss tolerant hybrid entanglement of light, Optica, 5, 1012.
- Guccione, et al. Connecting heterogeneous quantum networks by hybrid entanglement swapping, Sci. Adv., eaba4508

Bill Phillips: Concerning continuous variable quantum information—coherent states are not mutually orthogonal. Does this impose a fundament limit on fidelity of some operations, and does it matter?

If you are only interested in the orthogonality of the states, the coherent states with an amplitude of $\alpha=1$ obtained in our lab are already quite orthogonal, with an overlap of e-4 ~ 1.8%. We can thus reach a clean CV basis without the need to increase much more the size of the cat. Yet there are some protocols for which the orthogonality of the states is not a sufficient requirement, and in the case of a CV teleportation for example, the feed-forward necessary to recover the teleported state can only be performed efficiently for $\alpha >> 1$. Such an example is detailed in these references:

- Jeong, et al, Quantum-information processing for coherent superposition state via a mixedentangled coherent channel, PRA, 64, 052308.
- Park, et al., Quantum teleportation between particlelike and fieldlike qubits using hybrid entanglement under decoherence effects, PRA, 86, 062301.

3. Daniel Goncalves Romeu

In the 2D array as a perfect mirror, how does the the re ectivity depend on the motional temperature of the atoms?

DANIEL: Thermal motion can definitely affect the performance of the mirror array. Specifically, the mirror-like behavior arises from the interference of the fields emitted by the spatially ordered atoms. However, if these atoms are in different motional states, their emission becomes distinguishable, such that now the scattered fields do not interfere. In addition, heating increases the uncertainty in the atomic positions, which results in a weakening of the collective response as the array deviates from a perfect array of point-like scatterers. When thermal energy is high, these effects can heavily reduce the re ectance of the mirror. Although we do not expect to substantially heat our array (because we use weak fields), thermal effects must be considered in a real implementation.

Would it be possible to think of this potentially as a single photon transistor (where the presence of a single photon that interacts with the atoms and generates the Rydberg excitation can control the path of a single photon approaching the array)? Could this be done with photons in different modes, or in different frequencies to determine which photon is the "control" part of the switch?

DANIEL: While the definition of transistor can enclose further implications (for example, intensity-dependent control of the photon ux through the array), that is indeed the mechanism in our proposed switch: the storage of a single gate photon modifies the optical response of the whole array (from re ecting to transmitting) for a subsequent signal photon. In our case, we use two identical photons, where the gate photon is determined by the time sequence. In particular, we first store a gate photon, then send the signal photon, and finally coherently retrieve the gate photon at later times. Using photons in different modes can be problematic, as the efficiency of storage and re ectance depends on the light spatial mode. However, one can use photons with different frequencies, as long as the detuning of the auxiliary control field is properly modified between the storage and reflection events. Unfortunately, the switch re ects the photon back where it came from.

Would it be possible to create a switch that either transmits or reflects under 90 degrees?

Indeed, the reflection back into the exact same direction can be problematic from an experimental perspective. Fortunately, the perfect reflection in the array can also be achieved without requiring normal incidence, where the direction of an input Gaussian beam inciding with an angle follows the laws of classical specular reflection. Therefore, as long as the frequency conditions are satisfied and the angle is not too extreme, it should be possible to create a switch that transmits or reflects with an angle below 90 degrees and a with switch error similar to the one discussed in our work.

If one couples one atom to a waveguide or use non-gaussian em modes in free space one could have total reflection even with 1 atom. Would these "photonic" strategies help also for 2D arrays?

DANIEL: Aside from the error due to the finite array size, the unity reflection in a 2D array of point-like scatterers is theoretically limited by the different wavevector components ~k of the input field having different mirror resonance frequencies !~k. Since tightly focused Gaussian beams contain a wide range of wavevectors, it is impossible that all components simultaneously satisfy their frequency conditions for perfect reflection. This introduces an error that scales as $1/w_0^4$, where w_0 is the Gaussian beam waist. Within this context, perhaps one could obtain a better scaling by considering a different lattice structure yielding a potentially atter band, or a light mode with tailored wavevector components that further maximizes reflection.

Which would be the biggest experimental challenges in performing the experiment?

DANIEL: From the perspective of the mirror array, some of the main challenges are achieving near unity filling factor and reducing the thermal motion effects addressed in previous questions. On top of these, one needs that the Rydberg excitation remains stored throughout all the protocol, requiring techniques to prevent its accidental retrieval, decay or ionization. However, we would like to remain optimistic, as we are combining two physical ideas (mirror array and Rydberg blockade mechanism) that have already been implemented experimentally.

What is the conceptual difference between your proposed single-photon switch and the previous experiments on single-photon switches with unordered Rydberg atoms in dipole traps by the Vuletic group?

DANIEL: The main conceptual difference between our proposal and some of the extensive work of Prof. Vuletic's group in unordered Rydberg atoms is that we use an ordered atomic array, which guarantees efficient atom-light interactions without scattering into unwanted directions. Furthermore, we note that Prof. Lukin's group also has seminal work involving a mirror square array used to generate Schrödinger-cat states (quantum metasurface paper). However, our proposal is different in the atom that allows the switching between transmission and reflection, where any atom in the array can create an aperture.

What is the main difference between using the "standard" Rydberg EIT approach and your Rydberg dressing potential?

DANIEL: Phenomenologically, both Rydberg EIT (rEIT) and our Rydberg dressing approach are equivalent. Indeed, a similar switch could be created using the standard rEIT, where a transparent array becomes reflecing after the storage of a gate photon. The main difference is that the resulting mirror in the rEIT approach has one atom excited in the Rydberg state, which creates a singleatom aperture that reduces reflection. This is avoided in our Rydberg dressing approach, as the single-atom hole appears in the transmitting scenario and always lays within the larger aperture created by the dressing-induced potential.

Bill Phillips: How would things change if the atoms were not in a regular array, but in a disordered gas?

The coherent control and low dissipation in our proposed switch can be achieved thanks to the perfect reflection behavior of 2D arrays, which arises from the spatial periodicity in the array. Without said periodic structure, the mirror-like reflection is not possible and photons are scattered into random unwanted directions. While there have been proposals using disordered ensembles, dissipation becomes a major problem, limiting the performance of potential implementations such as photon-photon gates.