QSS47 - Vahid Sandoghdar - Questions & Answers

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When you look at the decoherence for your two level systems coupled to the environment, to what extent is there a focus in your research area on finding an environment where these are close to a theorist's two-level system (perhaps like your single-photon gun, for example), and to what extent are these imperfections actually a focus of the study, with the two-level system used a (spectroscopic) probe of the complex properties of the molecules/materials.

VAHID: Excellent question! For the past 20 years plus, we have worked with molecular crystal matrices in which the dye molecule of interest has a Fourier-limited linewidth at temperatures roughly below 2K. Exactly which system and which temperature happen to get you to this limit is not something that has a recipe, but there are a handful of systems that just work and are very robust. You can consult a very useful book from the early days of single-molecule spectroscopy in 1996 to see some of the variations: Single-Molecule Optical Detection, Imaging and Spectroscopy, edited by T. Basché , W.E. Moerner, M. Orrit, and U.P. Wild (VCH Verlagsgesellschaft, Weinheim, 1996). Single molecules were already shown in early times to be sensitive to pressure, electric field and phononic perturbations (see the book above). Indeed, recently we have become again interested in using single molecules as nanoscopic probes for their environments. You can see, e.g. Shkarin, et al, PRL 126, 133602 (2021). Also, the phononic coupling to the environment is very promising for future studies. For a theoretical example, please have a look at Gürlek, et al, arXiv:2104.01254 (2021).

VAHID: Can you derive from your work basic requirements needed (or basic principles) to turn a complex molecule into a perfect emitter?

Unfortunately not, but there are some general handwaving indicators. For example, a molecule that can bend about its bonds too much ends up having internal vibrations that reduce the branching ratio of the zero-phonon line. There is a huge literature on related topics in the physical chemistry community but a lot of the know-how and wisdom are burried in articles and books that are not easily accessible to someone with a quantum optics or atomic physics background. We are now trying to connect these two. An example is a recent theory paper: Reitz, et al, Phys. Rev. Res. 2 033270 (2020).

A finesse of 17k is not that high compared with some mirrors, what limits this number in your experiments, and is it surface roughness?

VAHID: Our finesse is essentially limited by the reflectivity of our mirrors, which is on purpose not chosen to be too high. As I mentioned in the lecture, we want to keep our Q as low as possible while achieving what we set out to achieve because it is a challange to lock a cavity in the cryostat. Having said that, at some point, the presence of the crystal itself will limit the Q or finesse. In any event, our surface roughness is not to blame yet.

It is possible to combine these systems with a photonic crystal cavity?

VAHID: In principle yes. However, you have to think about the desing details and whether you purturb the PC cavity. In other competing systems that use PC cavities such as quantum dots or diamond color centers, the PC is usually made of the host material, e.g. GaAs, Si, or diamond. Otherwise, adding a piece of an external object, even if nanoscopic, will at some point purturb the

PC cavity due to scattering. To get a bit deeper into this discussion, you can have a look at an older theory paper we wrote: Koenderink et al, PRL 95 153904 (2005) .

Since you were talking about coherence and decoherence, is it possible to obtain indistinguishable photons from a molecule?

VAHID: Because our molecules reach a Fourier-limited linewidth at liquid helium temperature, you can easily use them to generate indistinguishable photons. We demonstrated this for two separate molecules in Lettow et al, PRL 104 123605 (2010). This was demonstrated for a single molecule by A. Kiraz et al., Phys. Rev. Lett. 94, 223602 (2005) a few years earlier.

If you couple it to a plasmonic antenna, can indistinguishable photons be generated at room temperature?

VAHID: Coupling to plasmonic antenna can strongly shorten the spontaneous emission rate so that one could hope for it to dominate any other dephasing or nonradiative rate at room temperature. This limit has not been reached yet, but it is definitely something to strive for. You just have to keep in mind that with a plasmonic antenna alone, you will never reach a large quantum efficiency because the antenna has intrinsic material losses that quench your emitter in the near field. However, you can manage to have a substantial enhancement and maintain a quantum efficiency of the order of 70-80%. So to come back to your question, plasmonically-coupled indistinguishable photons at room temperature would be difficult, at least if you wanted them to be indisinguishable over a reasonabley long time.

When you look at the system of your two level system when you have photon bunching in transmission from the cavity, how would you write the quantum state of the transmitted light?

VAHID: One can set up a master equation following the open Jaynes-Cumming model. In the limit of infinite cooperativity and broad cavities ($\kappa \gg \gamma$) the wavefunction of the system tends to $|00\rangle - 2E^2/\kappa^2 |20\rangle$, where $|n_c\, n_e\rangle$ refers to product states of 'n_c' Fock number of cavity photons and 'n_e' number of emitter excitations. This is explained in the Supplementary Information (Eq. S13) of D. Wang, et al, Nat. Phys. 15 483 (2019).

Why are the two photons retained to some extent when passing through the cavity? Can we predict how many pairs of 2 photons should be scattered/measured?

VAHID: This is basically the result of a simple saturation effect: once the molecule is occupied with one photon, it cannot attend to any others that might be present in the cavity. As a result, single-photon states get filtered out. For a deeper discussion, you can consult some papers of the group of F. Laussy, e.g. Nat. Photonics 8, 550 (2014).

Looking at the array of plasmonic nanoparticles, do you envisage the possibility also to observe collective effects in the molecule emission?

VAHID: In principle, yes. But this would be technically quite challenging: you would have to couple a molecule to each nanopartifcle in its near field AND have all molecules have the same transition frequencies. We think that collective effects are more accessible if you couple many molecules to a single mode of a cavity, either in an open-cavity architecture or on a chip via nanoguides. You can consult Haakh et al, Phys. Rev. A. 94 053840 (2016) for the theory of such a system.

There are experiments that aim to modify not only radiative properties of molecules but even chemical reactions by using high-finesse cavities. What is your view on the possibilities, and are there applications of your work in that direction?

VAHID: So far, we have not worked with molecules that undergo any chemical modification. In fact, in our cryogenic line of work, we have been interested in molecules that are as photostable as possible. But, our cavity technology could certainly be employed to investigate systems like that.

What you have to keep in mind though is that a real high finesse means narrow lines and the molecules that have been studied in the context of chemical reactions don't have narrow lines. In fact, the cavities in those systems have quite low finesse values.

How important is mode-matching the incoming laser beam to the dipole radiation pattern of the molecule in determining your factor alpha in the extinction cross section. Does this matter also with a cavity?

VAHID: What we call alpha refers only to the properties of the molecule and how it interacts with its matrix. So in that sense, alpha does matter for work on cavities too: the lower alpha, the stronger your cavity coupling needs to be to achieve what you want. Mode matching is also important though, both in free-space focusing and for coupling to a cavity because what is not matched will not interact with the molecule.

If you were to illuminate the molecule with a fast light pulse, does the extinction cross section depend on the length of the pulse, i.e., if the interaction is faster than some of the decoherence processes, should we think of a time-dependent cross section?

VAHID: The extinction cross section that I quote assumes a resonance condition. In general, you have to multiply it by a Lorentzian function with the transition linewidth. So, now you see that if your pulse is broader than the molecular transition linewidth, you don't couple well.

Apart from the fast processes leading to the dephasing rate you mentioned, are there also resonance frequency fluctuations on longer time scales? Is this relevant in situations where long-time illumination is required?

VAHID: Depending on the combination of the dye molecule of interest and its matrix, there might well be spectral instabilities.Of course, if the temperature of your cryostat is not stable, you would see spectral instabilities, but this is a technical problem. In „good" systems, spectral diffusion can be very small, even over hours, within about a linewidth. In polymers, this is a huge problem.