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Observation of Two-Photon Interference Using the Zero-Phonon-Line Emission of a Single Molecule

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Abstract. We report the results of coincidence counting experiments at the output of a Michelson interferometer using the zero-phonon-line emission of a single molecule at 1.4 K. Under continuous wave excitation, we observe the absence of coincidence counts as an indication of two-photon interference. This corresponds to the observation of Hong-Ou-Mandel correlations.

1. Two-Photon Interference

Two photons which are sent to two different input channels of a 50/50 beam-splitter will interfere depending on their indistinguishability. Ideally indistinguishable photons will exhibit the bunching behavior, i.e. they will leave the beam splitter through the same output channel [1]. Apart from being a major demonstration of the quantum theory of light, this phenomenon has so far proven to be very suitable for applications in quantum information science. It is conceivable to perform efficient quantum computation using only indistinguishable single photons, linear optical elements and ideal photon counters [2].

The availability of true single photon sources which should provide indistinguishable single photons on demand is a major practical requirement of the linear optics quantum computation scheme. Photons generated in a parametric down conversion process have been frequently used as such sources [3]. However, in this source, the number of generated photon pairs is governed by Poissonian statistic. Thus, experiments using parametric down conversion are bound to be performed at low intensities.

Single photon sources based on single two-level emitters promise to overcome this inherent limitation [4, 5, 6]. These single photon sources can emit single photons with a sub-Poissonian photon statistics provided a large collection efficiency. Only very recently, the coherence properties of the emitted single photons have been investigated. Impressive demonstrations of indistinguishability have been given for photons emitted by single quantum dots [7] and single trapped atoms [8]. Single molecules are an attractive alternative to both single quantum dots and single trapped atoms. Among the available systems, only single molecules can provide large photon coherence lengths together with large observation times. We have readily observed photon coherence lengths close to 4.9 ns [9] for stable single molecules which can fluoresce over 20 days without photobleaching at cryogenic temperatures [10]. In the following, we report on Hong-Ou-Mandel experiments using the zero-phonon-line (ZPL) emission from a single

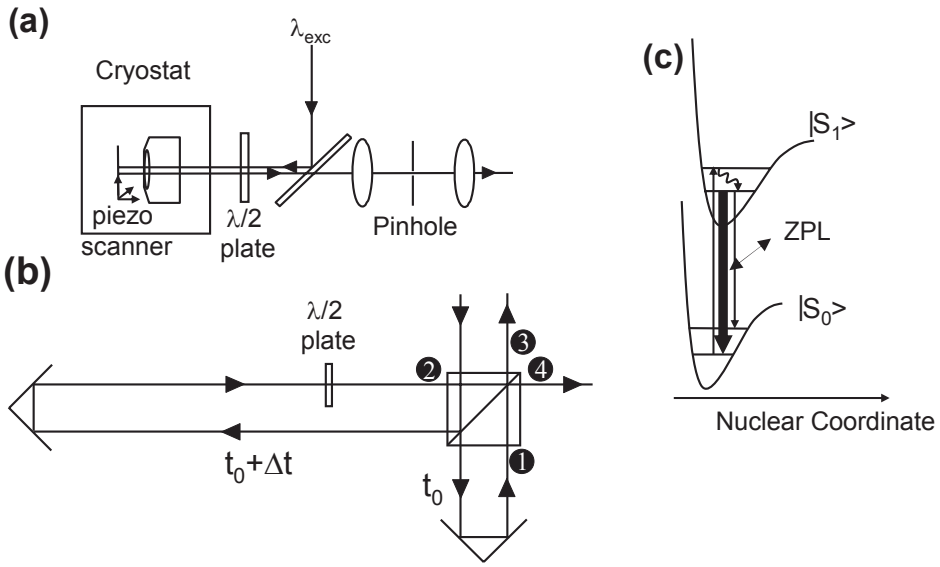

Figure 1. (a) Low temperature confocal microsocope. (b)Michelson interferometer. (c) Vibronic excitation scheme.

terrylenediimide (TDI) molecule. As a result of these experiments, we demonstrate the two-photon interference phenomenon [11].

2. Experimental Setup & Sample Preparation

A home-built low temperature confocal microscope setup was used in the experiments (Figure 1(a)). In this setup, the beam from a single mode dye laser was reflected off a dichroic mirror, passed through a $\lambda/2$ plate before being focused onto the sample with an aspherical lens (NA=0.55), which is also used for collection. A piezo scanner was used to select single molecules from different spatial positions. After transmision through the $\lambda/2$ plate and the dichroic mirror, the collected fluorescence was focused onto a pinhole (75 μm diameter) and sent to a home-built Michelson interferometer depicted in Fig. 1(b). Alternatively the signal was dispersed with a 46 cm monochromator before detection with a charge coupled device detector. A spectral resolution of 30 GHz was obtained in the spectra taken with the monochromator and the CCD.

The experiments were performed on terrylenediimide (TDI) molecules [12] highly diluted in hexadecane (HD) matrix. Hexadecane forms a Shpol'skii matrix which provides an ordered, partly crystalline surrounding for the TDI molecules. The samples were prepared by adding hexadecane to a solution of TDI in $CHCl_3$. $CHCl_3$ and O_2 were removed by several freeze and thaw cycles using liquid N_2 and intermediate evacuation steps. After this, the TDI/hexadecane solution was saturated with Ar. A drop of this solution was then quickly inserted into the precooled cryostat. Finally, the sample was cooled down to 1.4 K in a liquid He bath.

The technique of vibronic excitation was used to excite single TDI molecules during the experiments [13] (Fig. 1(c)). At cryogenic temperatures, due to their short lifetimes (∼1-10 ps), the vibrational states of single dye molecules are generally very broad. The usable absorption width is further increased to tens of cm^{-1} in systems with a strong electron-phonon coupling due to intense phonon sidebands. Hence, when a vibrational state is excited and ZPL emission is detected in a spectrally resolved manner, large spectral jumps (up to 80 cm^{-1} [13]) can be observed. Vibronic excitation also allows for the detection of the ZPL which is indispensible for the experiments reported in this proceeding.

3. Experimental Results

A Michelson Interferometer is typically used in experiments performed to observe two-photon interference using the emission collected from one single photon source [7, 8]. Under cw excitation, a path length difference which is larger than half of the coherence length of the emitted photons should be selected. This ensures that two independent photons will simultaneously arrive at two inputs of the beam splitter with a certain probability. The signature of two-photon interference is then revealed as the lack of coincidence counts at the output of the beam splitter.

In contrast to the recently reported two-photon interference experiments using single quantum dots [7] and single trapped atoms [8], we have employed continuous wave (cw) excitation in our experiment. As depicted in Fig. 1(b), we have implemented a rotatable $\lambda/2$ plate in the longer arm of the interferometer in order to achieve parallel or orthogonal polarizations in channels 1 and 2. Considering parallel polarizations, in the limit of large Δt ($\Delta t > 1/\gamma$) and small τ ($\tau < \Delta t$), a coincidence counting experiment between channels 3 and 4 reveals the normalized second-order coherence function [14]:

$$g^{(2)}_{34\parallel}(\tau) \;=\; \frac{1}{2}\left(g^{(2)}(\tau)+1\right) - \frac{\sin^2\theta\cos^2\theta}{\cos^4\theta+\sin^4\theta}\left|g^{(1)}(\tau)\right|^2 . \qquad (1)$$

$\gamma = \Gamma_{spon}/2 + \gamma_{pure}$ is the total dephasing rate of the ZPL including spontaneous emission (Γ_{spon}) and dephasing due to other sources (γ_{pure}). In this equation $g^{(1)}(\tau)$, $g^{(2)}(\tau)$ correspond to the normalized first-order and second-order coherence functions of the ZPL emission of a single molecule respectively. Transmission and reflection in the beam-splitter are noted as $\cos^2\theta$ and $\sin^2\theta$ respectively.

For mutually orthogonal polarizations, photons at both input channels are completely distinguishable, and no effect of two-photon interference should be observed. For this case, the result of a coincidence experiment between channels 3 and 4, reveals:

$$g^{(2)}_{34\perp}(\tau) \;=\; \frac{1}{2}\left(g^{(2)}(\tau)+1\right) . \qquad (2)$$

Examplary solutions of $g^{(2)}_{34}(\tau)$ for both mutually parallel (solid) and orthogonal (dashed) polarizations are depicted in Fig. 2(e). In the solid curve, the signature of two-photon interference is the absence of coincidence events around zero delay time.

For the two-photon interference experiments, the confocal microscope was equipped with a piezo scanner in order to select single molecules from different spatial positions in the sample. After being filtered with a narrow band interference filter (FWHM=1nm, angle tunable between 645-670 nm), ZPL fluorescence from a single TDI molecule was directed into the home-built Michelson interferometer. The path length difference between the two arms of the interferometer was equivalent to a temporal delay of $\Delta t = 4.6\ ns$. One of its arms was equipped with a rotatable $\lambda/2$-plate. The output signals at the beam splitter were detected using APDs in a Hanbury-Brown and Twiss configuration. The APD outputs were sent to a time to amplitude converter (TAC) as start and stop pulses. After adding an electronic delay on the stop pulse, the output of the TAC which was stored in a multi channel analyzer (MCA) revealed the second order correlation function ($g^{(2)}_{34}(\tau)$). A temporal resolution of 420 ps was achieved.

Fig. 2(a) depicts the results of coincidence measurements for the case when the $\lambda/2$-plate introduced no polarization change in the long arm of the interferometer (parallel polarizations). At the zero delay position, the value of $g^{(2)}(0)$ becomes 0.4. Fig. 2(b) depicts the result of the coincidence detection experiment when we have rotated the polarization of the fluorescence in the long interferometer arm by 90°. Clearly, the resulting correlation curve exhibits higher $g^{(2)}$ values around zero delay time. The normalized difference between the two curves is shown in Fig. 2(c). In this figure the dip around zero delay time constitutes the proof for the observed

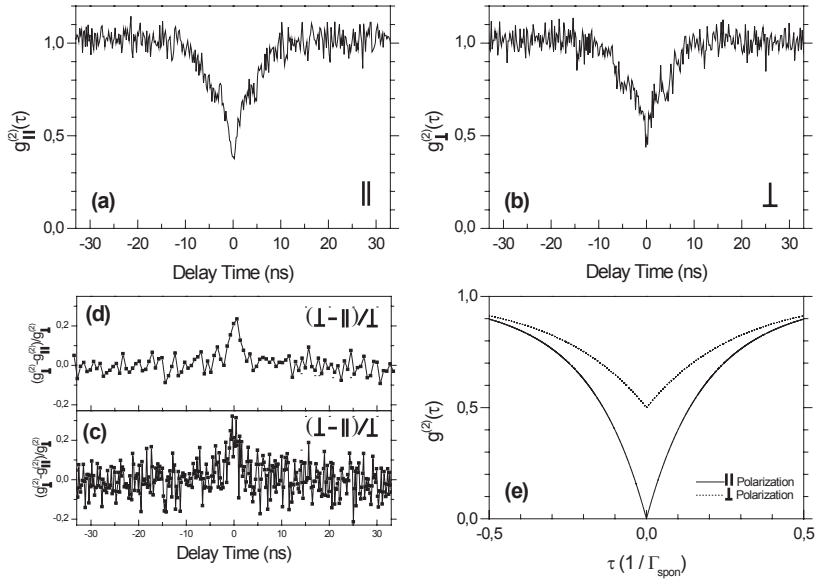

Figure 2. Measured normalized second-order photon correlation functions for parallel (a) and orthogonal (b) polarizations. Binning of the data is 2 in a and b. Difference between the correlation functions $g_{\perp}^{(2)}$ and $g_{\parallel}^{(2)}$ normalized to $g_{\perp}^{(2)}$ with a binning of 2 (c) and 7 (d). Excitation is at 605 nm with an intensity of 1 mW in a-d. (e) Solution of $g_{34}^{(2)}(\tau)$ for large Δt ($\Delta t \gg 1/\gamma$) and small τ considering parallel and ortohogonal polarizations. Parameter values are $W_P = 0.25\Gamma_{spon}$, $\gamma_{dep} = 0.3\Gamma_{spon}$, and $\theta = \pi/4$. W_P is the effective pumping rate of the two-level emitter.

Hong-Ou-Mandel correlations. The dip is clearly visible in Fig. 2(d) where a binning of 7 (time bin = 658 ps) was used. When compared to the photon coherence times of a few nanoseconds, the time resolution in Fig. 2(d) is still sufficient for the observation of two-photon interference. Furthermore, we have observed the normalized difference between the data of Fig. 2(a) (2(b)) and another measurement with parallel (orthogonal) polarizations [11]. In contrast to Fig. 2(c), both of the cases revealed no signal above the noise level at zero delay.

The observed $g^{(2)}(0) = 0.4$ for parallel polarizations corresponds to a photon indistinguishability of at least ~ 0.2 instead of the ideal value of 1. Two reasons can be responsible for this relatively low level of indistinguishability. Firstly, imperfections in the mode matching of our interferometer are a source of contrast reduction. Because of its higher throughput, a conventional Michelson interferometer was preferred over a setup using a single mode glass fibre in our experiment. Secondly, spectral jumps of the molecule could deteriorate the contrast. If these jumps occur on the time scale of the excited state lifetime, they will give rise to dephasing and a corresponding line broadening. While we did not determine the ZPL emission linewidth of this molecule, a fit to the correlation measurement in Fig. 5(a) reveals $\Gamma_{spon} \sim 1/(3\ ns)$ and $\gamma_{pure} \sim 1/(4\ ns)$, hence a photon coherence time of $\sim 2.5\ ns$.

4. Conclusions & Outlook

In conclusion, we have observed the two-photon interference phenomenon using the zero-phonon line (ZPL) emission of a single molecule at cryogenic temperatures. Using a Michelson interferometer, a photon indistinguishability of at least ~ 0.2 was observed as compared to the ideal value of 1. Our results yield single molecules as an attractive alternative to single

InAs quantum dots and trapped atoms for applications in linear optics quantum information processing. The two-photon interference experiments presented in this proceeding can easily be extended to include a pulsed excitation scheme. Although pulsed excitation scheme is not necessary for the demonstration of two-photon interference, it will be important for practical applications. Our approach offers new possibilities for performing linear optics quantum information experiments like those reported in [7, 15, 16] using larger coherence times.

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References

[1] P. Grangier. *Nature*, 419:577, 2002.
[2] E. Knill, R. Laflamme, and G. J. Milburn. *Nature (London)*, 409:46, 2001.
[3] C. K. Hong, Z. Y. Ou, and L. Mandel. *Phys. Rev. Lett.*, 59:2044–2046, 1987.
[4] C. Brunel, B. Lounis, P. Tamarat, and M. Orrit. *Phys. Rev. Lett.*, 83:2722, 1999.
[5] P. Michler, A. Kiraz, C. Becher, W. V. Schoenfeld, P. M. Petroff, Lidong Zhang, E. Hu, and A. Imamoğlu. *Science*, 290:2282, 2000.
[6] Axel Kuhn, Markus Hennrich, and Gerhard Rempe. Deterministic single-photon source for distributed quantum networking. *Phys. Rev. Lett.*, 89:067901, 2002.
[7] C. Santori, D. Fattal, J. Vuckovic, G. S. Solomon, and Y. Yamamoto. *Nature*, 419:594–597, 2002.
[8] T. Legero, T. Wilk, M. Hennrich, G. Rempe, and A. Kuhn. *Phys. Rev. Lett.*, 93:070503, 2004.
[9] A. Kiraz, M. Ehrl, C. Bräuchle, and A. Zumbusch. *Appl. Phys. Lett*, 85:920, 2004.
[10] F. Kulzer, S. Kummer, R. Matzke, C. Bräuchle, and Th. Basché. *Nature*, 387:688, 1997.
[11] A. Kiraz, M. Ehrl, T. Hellerer, Ö. E. Müstecaplıoğlu, C. Bräuchle, and A. Zumbusch. *Phys. Rev. Lett*, 94:223602, 2005.
[12] S. Mais, J. Tittel, T. Basché, C. Bräuchle, W. Göhde, H. Fuchs, G. Müller, and K. Müllen. *J. Phys. Chem.*, 101:8435, 1997.
[13] A. Kiraz, M. Ehrl, C. Bräuchle, and A. Zumbusch. *J. Chem. Phys.*, 118:10821–10824, 2003.
[14] A. Kiraz, M. Atatüre, and A. Imamoğlu. *Phys. Rev. A*, 69:032305, 2004.
[15] D. Fattal, K. Inoue, J. Vuckovic, C. Santori, G. S. Solomon, and Y. Yamamoto. *Phys. Rev. Lett.*, 92:037903, 2004.
[16] D. Fattal, E. Diamanti, K. Inoue, and Y. Yamamoto. *Phys. Rev. Lett.*, 92:037904, 2004.