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## Development of a parametric down-conversion source for two-photon absorption experiments

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# Development of a Parametric Down-Conversion Source for Two-Photon Absorption Experiments

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## ABSTRACT

We describe a fiber-coupled parametric down-conversion (PDC) source designed for entanglement-enhanced two-photon absorption experiments. The key feature of the source is a narrowband ( $\sim 1$  MHz) UV diode pumping laser which can be tuned to match the energy of the  $5S_{1/2}$  to  $5D_{5/2}$  two-photon transition in Rubidium. The weak narrowband pumping beam is delivered to the PDC crystal through a single-mode fiber, which allows the source to be pre-aligned with a much stronger broadband auxiliary pump laser. The motivation for this PDC source lies within the context of Linear Optics Quantum Computing and Quantum Zeno Gates.

**Keywords:** parametric down-conversion, two-photon absorption, quantum Zeno Gates

## 1. INTRODUCTION AND MOTIVATION

It has recently been shown<sup>1</sup> that the failures inherent in probabilistic Linear Optics Quantum Computing (LOQC) logic gates<sup>2</sup> can be overcome by using the quantum Zeno effect. This results in the possibility of implementing deterministic “Zeno Gates”, which may offer a number of advantages for quantum information processing with single-photon qubits.<sup>1</sup> One promising approach to implementing Zeno Gates involves the use of strong two-photon absorption at single-photon intensity levels.<sup>1</sup> In these proceedings, we describe a special parametric down-conversion (PDC) source designed for use in two-photon absorption experiments related to Zeno Gates.

The large two-photon absorption cross sections that will be required for Zeno Gates are difficult to achieve under ordinary circumstances.<sup>3</sup> However, it is well known that two-photon absorption probabilities can be significantly enhanced by reducing the mode-volume associated with the photons,<sup>4</sup> and by exploiting any frequency-entanglement of the photon-pairs to be absorbed (see, for example,<sup>5–11</sup>). Although our preliminary Zeno Gate experiments will utilize both of these features, the focus of the present work is related to the latter. The goal is the development of a PDC source that essentially maximizes the frequency-entanglement of the down-converted pairs by using a narrowband pump laser.

## 2. TWO-PHOTON ABSORPTION WITH FREQUENCY ENTANGLED PHOTONS

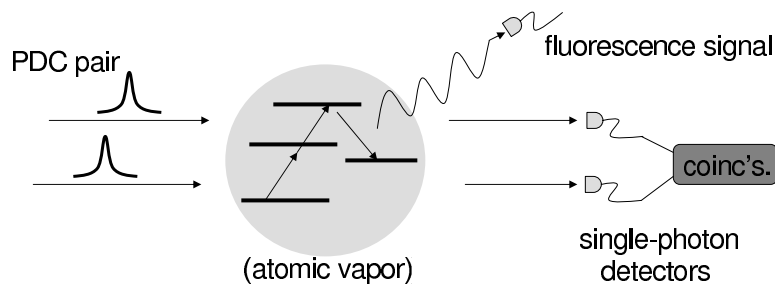
There is a long history of interest in two-photon absorption with nonclassical states of light. The pioneering experiments of Georgiades *et al.*<sup>5,6</sup> used squeezed-vacuum states to successfully observe a two-photon absorption rate that grew linearly with intensity,<sup>7,8</sup> rather than quadratically, as would be expected by using classical light sources. Very roughly speaking, this nonclassical effect can be understood by the fact that an ideal squeezed-vacuum state is described by a superposition of even-numbered photon terms, so increasing the intensity of the source simply increases the number of photon pairs in the beam.

More recently, Dayan *et al.* have conducted a series of very illustrative experiments that focused on the energy-time entanglement of the photon pairs being absorbed.<sup>9–11</sup> As emphasized by Dayan *et al.* in reference,<sup>9</sup> strong two-photon absorption appears to have conflicting requirements: the two photons must arrive at an atom at the same time (ie. they should be short duration “broadband wavepackets”), and the sum of their energies must equal that of the atomic transition (ie. they should be well-defined “narrowband wavepackets”).

Remarkably, these requirements can be circumvented by exploiting the energy-time entanglement of photon pairs produced via PDC. In typical PDC experiments, the individual photons of a pair can be extremely broadband (often 10's or even 100's of nm), but the *sum* of their frequencies must equal that of the pumping beam. Consequently, if the frequency of the pumping beam is tuned to match the relevant atomic transition, entanglement-enhanced two-photon absorption can occur, which leads to a number of interesting phenomena.<sup>12</sup>

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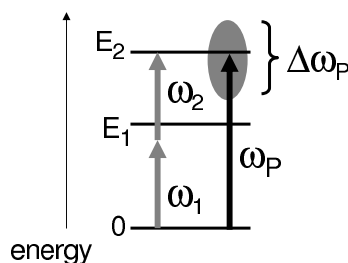
**Figure 1.** Basic idea of two-photon absorption experiments using the PDC source. The goal is to realize a two-photon absorption cross section that is large enough to create an observable reduction in the coincidence counting rate between two single-photon detectors. This represents a different regime than earlier work which essentially used large numbers of entangled-pairs to generate a weak fluorescence signal from the spontaneous emission of two-photon excited atoms.<sup>9</sup> The large cross sections needed to produce an observable dip in the coincidence rate are required for quantum Zeno Gate applications.<sup>1</sup>

### 3. COINCIDENCE COUNT RATE REDUCTION VS. FLUORESCENCE SIGNAL

In earlier work, large numbers of entangled photon pairs passed through an atomic medium, and two-photon absorption was signalled by a fluorescence signal generated through spontaneous emission from the excited atoms. (This technique is also used in typical Doppler-free two-photon absorption experiments with “classical” beams: see, for example,<sup>13</sup>). The advantage of this arrangement is that the physics of entanglement-enhanced two-photon absorption can be studied even if the absorption cross section is relatively small.

In contrast, our PDC source is being developed for experiments involving small numbers of entangled photon pairs, but very large two-photon absorption cross sections. As illustrated in Figure 1, the goal is to achieve a situation in which every PDC pair has a substantial probability of being absorbed, which would lead to an observable reduction in the coincidence counting rate between two single-photon detectors.

The need to observe an actual removal of pairs, rather than a fluorescence signal, places strict requirements on the design of the PDC source. As shown in Figure 2, the sum of the down-converted photons’ energies is equal to that of the pump (eg.  $\hbar\omega_1 + \hbar\omega_2 = \hbar\omega_p$ ), but that value is “smeared” by the bandwidth of the pump. Consequently, if the bandwidth of the pump is much wider than the bandwidth associated with the two-photon transition, there will be a decrease in the overall two-photon absorption rate, and a corresponding increase in the background (eg. noise) in the coincidence counting rate. For experiments based on the fluorescence signal this background is irrelevant, which allows the use of a relatively broadband pump.<sup>9</sup>



**Figure 2.** Illustration of the need for a narrowband pump laser for the PDC source. The down-converted photons’ energies sum up to that of the pump, but that value is “smeared” by the bandwidth of the pump ( $\Delta\omega_p$ ). The probability of two-photon absorption will be reduced if  $\Delta\omega_p$  is broader than the linewidth of the two-photon transition.

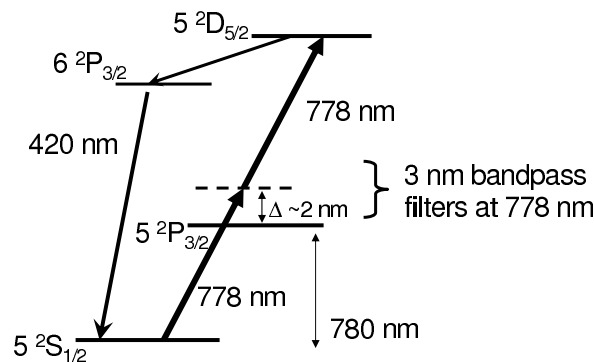
In order to see a large signal-to-noise ratio in the expected “dip” of coincidence rate, our source requires a pump laser with a bandwidth much less than the broadened linewidth of the atomic transitions of interest ( $< 0.5$  GHz). Most typical PDC pump lasers, however, have bandwidths that are orders of magnitude wider. For example, frequency-doubled Ti-Sapph. pulses can have a bandwidth of several nm’s (eg. THz), and even good low-power UV diode lasers can have a bandwidth on the order of 0.1 nm (eg. 100 GHz). A key feature of our PDC source is the use of a grating-stabilized external-cavity diode laser pump (Toptica Model DL-100), which has a bandwidth of  $\sim 1$  MHz and can be tuned to the appropriate UV wavelength for planned Zeno Gate experiments in rubidium vapor.

#### 4. RUBIDIUM CONSIDERATIONS

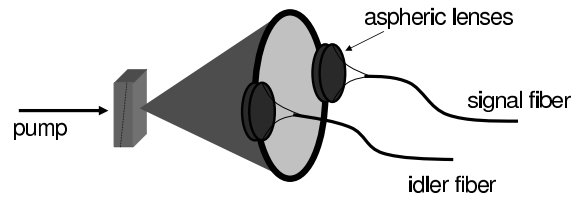
Our PDC source is designed for two-photon absorption experiments using the well studied  $5S_{1/2} \rightarrow 5D_{5/2}$  two-photon transition in Rb (see Figure 3). In this transition, two photons at 778 nm experience a relatively large two-photon absorption cross-section because of a relatively small ( $\Delta \sim 2$  nm) detuning from the  $5P_{3/2}$  intermediate state. Because of the strong cross-section, and the availability of narrowband diode lasers at 778 nm, this transition is often used in precision Doppler-free two-photon spectroscopy experiments<sup>13</sup> for atomic clocks.

In accordance with Figure 3, our PDC source uses a 389 nm pump to generate twin photons at 778 nm. The bandwidth of the down-converted photons can be restricted by using interference filters, and is a crucial parameter for optimizing the signal-to-noise ratio in the coincidence-counting experiments. It is desirable to have the photons broadband (eg. “short wavepackets”), but if the bandwidth is too wide there exists two-photon amplitudes that do not leverage the small detuning from the intermediate  $5P_{3/2}$  state. Once again this results in a smaller two-photon absorption cross section, and a corresponding increase in background coincidence counts. In addition, it is desirable to minimize single-photon loss by staying detuned from the strong 780 nm ground state transition. Our preliminary estimates indicate that interference filters centered at 778 nm, with a bandwidth of about 3 nm, should be optimal.

As shown in Figure 4, the twin photons at 778 nm are generated by non-collinear Type-I PDC and collected into single-mode fibers, which are required for integration with the various mode-reducing devices to be used in the initial two-photon absorption experiments. We use a fiber-coupling strategy based on the methods developed by Kurtsiefer *et.al*,<sup>14</sup> and attempt to maximize the number of pairs collected into the fibers. This requires moving the collection lenses as close to the crystal as possible to maximize the percentage of the 778 nm “ring” coupled into the fibers, and utilizing a relatively small diameter pumping beam to increase the collection efficiency.<sup>15</sup>



**Figure 3.** Energy level diagram for the  $5S_{1/2} \rightarrow 5D_{5/2}$  two-photon transition in Rb. Two photons at 778 nm experience a relatively large two-photon absorption cross section due to a small detuning ( $\Delta \sim 2$  nm) from the intermediate  $5P_{3/2}$  state. The use of interference filters with a 3 nm bandwidth (centered at 778 nm) is expected to increase the signal-to-noise ratio in our initial two-photon absorption experiments.



**Figure 4.** Overview of a fiber-coupled type-I PDC source. The PDC crystal emits a “cone” of light over the wavelength range of interest. In order to increase the number of entangled photon pairs collected into the fiber, the lenses are moved as close to the PDC crystal as possible. Roughly speaking, this maximizes the percentage of the “ring” that is imaged into the signal and idler fibers. In this scenario, a relatively small pump beam diameter also increases the number of collected pairs.

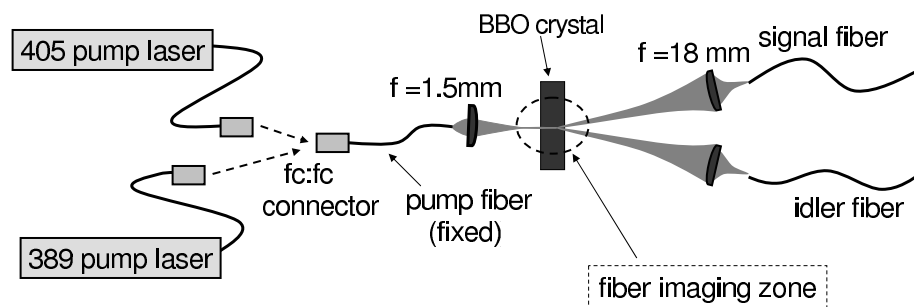
## 5. FIBER-COUPLED PUMPS FOR PDC

The Toptica DL-100 pump laser has an output power of less than 1 mW at 389 nm, which makes initial alignment of the fiber-coupled system shown in Figure 4 difficult. To overcome this problem, we use a single-mode fiber to deliver the pump beam to the PDC crystal. As shown in Figure 5, this allows us to pre-align the system using a much stronger ( $\sim 10$  mW) broadband diode pump laser at 405 nm.

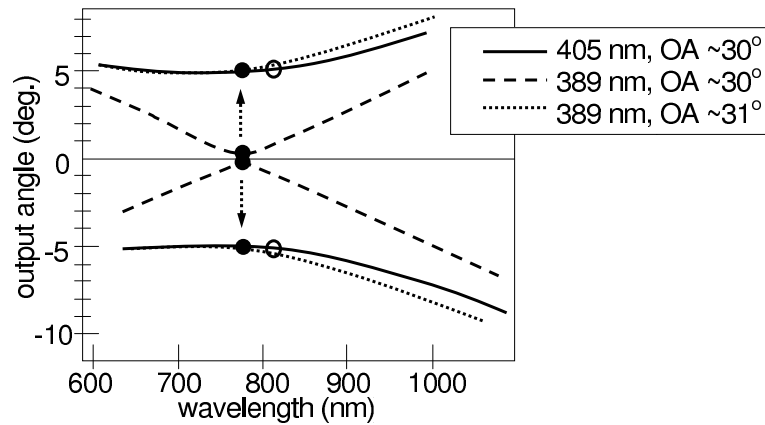
The basic idea is to use the strong signal generated by the 405 nm pump to easily optimize the system for collection of degenerate 810 nm PDC pairs. This ensures correct mode-matching (eg. fiber-to-fiber imaging) between the signal and idler fibers, and the pump fiber. Once the fiber and lens positions are optimized and fixed, the 405 nm pump laser is replaced with the 389 nm laser (which has been fiber coupled) using a simple fc:fc fiber connector.

Because the phase-matching conditions are slightly different for a 405 nm vs. 389 nm pump beam, the photon pairs at 778 nm emerge from the PDC crystal at a different angle than the 810 nm photons used to align the system. Consequently, the phase-matching conditions need to be adjusted by tilt-tuning the crystal until the 778 nm pairs are collected by the fibers.

Figure 6 illustrates the idea behind this tilt-tuning procedure. The “tuning curves” shown in the figure are derived from the phase-matching conditions, and represent the angles at which the PDC photons exit the crystal



**Figure 5.** Fiber-coupled pump beams for PDC. A strong ( $\sim 10$  mW) fiber-coupled auxiliary pump laser (at 405 nm) provides a bright PDC signal that is used to pre-align the fiber-collection system. Once the signal and idler collection fibers are optimally mode-matched to the pump fiber, the pump fiber and its focusing lens are fixed in position. At this point, the much weaker ( $\sim 100$   $\mu$ W) fiber-coupled narrowband pump (at 389 nm) is connected to the pump fiber. Because the fiber imaging zone is already optimized, the only adjustment needed is a known tilt of the BBO crystal to account for the slightly different phase-matching conditions of a 405 nm vs. 389 nm pump. The use of “fc” connectorized pump fibers allows rapid and easy switching between pump lasers.



**Figure 6.** Tuning curves calculated from PDC phase-matching conditions in BBO with a 405 nm pump, and a 389 nm pump. The plots show the angle at which the PDC photons leave the BBO crystal as a function of their wavelengths, for a given pump wavelength and optic axis (OA) orientation. The upper half of the plot (positive angles) corresponds to the signal photons, while the lower half (negative angles) correspond to idler photons. As described in the text, the open and solid circles denote degenerate wavelength photon-pairs of interest used for alignment and experiments.

as a function of their wavelength. The two solid curves show the situation with the 405 nm auxiliary laser, and the crystal oriented with its optic axis at  $30^\circ$  relative to the pump direction. As indicated by the open circles, this results in the 810 nm signal and idler pairs exiting the crystal at roughly  $5^\circ$ , which is the nominal “cone angle” defined by the geometry of the various optical elements in our setup.

When the 405 nm pump is replaced by the 389 nm pump (large-dash line), the 778 nm signal and idler photons of interest (denoted by solid circles) exit the crystal at roughly  $0.5^\circ$  and are not coupled into the fibers, which still sit at  $5^\circ$ . However, as the crystal is tilted away from the pump, the 778 nm pairs exit at increasing angles; finally reaching the  $5^\circ$  value required for fiber coupling (small-dash line) when the optic axis is oriented at  $31^\circ$ . In practice, the crystal is simply tilted in the correct direction until the 778 nm pairs are found.

The fiber-coupled pump at 389 nm currently delivers approximately  $100 \mu\text{W}$  of power to a 0.7 mm thick BBO crystal. Using standard 10 nm bandpass filters (centered at 780 nm), this results in a coincidence counting rate (between fiber-coupled single-photon counting modules; Perkin Elmer SPCM-AQR15’s) on the order of 10 pairs per second. Although relatively small, this count rate is expected to be adequate for our initial two-photon absorption experiments. Using custom 3 nm bandwidth interference filters at 778 nm (as described in Section 4) is expected to result in the detection of a few pairs per second and, based on phase-matching and fiber-coupling considerations, that rate should not vary significantly with pump wavelength scans over the range of interest (a few GHz).

## 6. SUMMARY

In summary, we have developed a fiber-coupled PDC source for use in entanglement-enhanced two-photon absorption experiments.<sup>5–11</sup> The source differs from conventional PDC sources by using an extremely narrowband ( $\sim 1$  MHz) tunable UV diode laser as a pump, and delivering the pump to the PDC crystal through a single-mode fiber. The source is designed for the goal of observing of an actual reduction in the pair detection rate (rather than a two-photon absorption fluorescence signal) after passing through a two-photon absorbing medium.

Our initial experiments will involve propagating the frequency-entangled pairs through tapered optical fibers in rubidium vapor. Our group currently has the capability of producing low-loss tapered fibers with diameters of less than 500 nm, which can be used to effectively reduce the mode-volume associated with the photon pairs. Subsequent experiments may involve the use of a holey-fiber microcavity to reduce the mode-volume.<sup>16</sup> The

goal is to combine the frequency-entanglement and mode-volume reduction to enhance the two-photon absorption cross-section to the point needed for Zeno Gate experiments.<sup>1</sup>

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