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Few-Photon All-Optical Modulation in a Photonic Band-Gap Fiber

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We demonstrate 25% all-optical modulation with <20 photons, i.e., a few attojoules of energy, using non-degenerate two-photon absorption in rubidium atoms confined to a hollow-core photonic band-gap fiber. An attenuation of up to 3 dB is induced on an optical field with a switching energy density of less than one photon per $(\lambda^2/2\pi)$. We show that the temporal response of the system is determined by the 5-ns transit time of the atoms across the optical mode of the fiber, which results in a modulation bandwidth up to 50 MHz.

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The ability to control light with light at ultralow powers has been a major avenue of research in photonics, and it is critical to demonstrate such effects near the single-photon level for quantum information applications [1]. Consequently, there is considerable interest in developing systems with high optical nonlinearities in geometries that enhance the light intensity by tight confinement of the optical mode. Various nonlinear and quantum-optical techniques are being explored for the realization of efficient all-optical switches [2–5], all-optical quantum computing gates [6, 7], and quantum non-demolition (QND) measurements [8–10].

Alkali vapors such as rubidium (Rb) have been used extensively for light-matter interactions due to the large cross-section per atom, well-defined energy level structure, long interaction times, and the large optical depths (ODs) that can be achieved [2–5, 11]. Optical waveguides such as photonic band-gap fibers (PBGFs) with a hollow core [see Fig. 1] allow various gases to be injected into the core and interact with single-mode optical fields [12, 13]. This architecture confines both the atoms and photons to a small transverse area, which permits weak fields to interact strongly with the atoms over a length that is much larger than the Rayleigh diffraction length for a focused beam of a similar spot size. Rb-PBGF systems have been shown to provide large ODs [14] and strong nonlinearities at low-light levels [5, 15]. However, the tight confinement of thermal atoms and photons give rise to additional spectroscopic features such as transit-time broadening that must be taken into account [16].

Two-photon absorption (TPA) is the simultaneous absorption of either two photons from a single beam of light (degenerate) or two single photons from two beams (non-degenerate) and results in a resonant transition from the ground state to an excited state. Many interesting two-photon processes and their applications, such as all-optical switching [17] and generation of single photons [18], require a significant TPA at low power levels. For example, it has been shown that the quantum Zeno effect can be used to implement optical logic gates for classical and quantum computing, where the Zeno effect is produced using a strong TPA medium [19]. Alkali

atoms such as Rb can have relatively large two-photon cross-sections due to near-resonant enhancement and the high oscillator strengths of their transitions. Nevertheless, TPA experiments in bulk alkali vapor cells require relatively high powers to generate measurable effects [20]. Waveguide geometries, such as hollow-core PBGFs filled with an alkali vapor or tapered optical fibers with an ambient thermal vapour of alkali atoms, can enhance nonlinear interactions greatly and significantly reduce the threshold for observing TPA [21–23].

Here we describe the observation of large TPA at sub-nanowatt power levels at the $^{85}\text{Rb } 5S_{1/2} \rightarrow 5D_{5/2}$ two-photon transition using a near-resonant, non-degenerate scheme in our Rb-PBGF system. This system demonstrates that as few as 20 photons can induce appreciable all-optical modulation on another beam and that it can be used to explore novel classical and quantum nonlinear effects at ultralow powers. In addition, we measure a fast response time of a few nanoseconds that can yield large modulation bandwidths.

The relevant energy levels of Rb and the two-photon level scheme are shown in Fig. 1. An atom in the $5S_{1/2}$ ground state can simultaneously absorb one photon at 780.2 nm (control) and another at 776 nm (signal) to make a resonant transition to the $5D_{5/2}$ excited state. By using a counter-propagating configuration, one can eliminate Doppler broadening in the TPA signal [20]. A small fraction of the excited atoms can decay back to the ground state through the $6P_{3/2}$ level emitting blue fluorescence. In the confined geometry here, most of the excited atoms collide with the walls of the fiber and decay non-radiatively [21]. The imaginary part of the third-order susceptibility corresponding to TPA [24] is given by $\chi^{(3)} = (N\mu_1^2\mu_2^2)/(\epsilon_0\hbar^3\Delta^2\gamma)$, where N is the atomic number density, μ_1 and μ_2 are the transition dipole moments for the $5S_{1/2} \rightarrow 5P_{3/2}$ and $5P_{3/2} \rightarrow 5D_{5/2}$ transitions, Δ is the detuning from the intermediate $5P_{3/2}$ level, and γ is the homogeneous decay rate from the $5D_{5/2}$ level. In the Rb-PBGF system, γ is determined by the transit time of the atoms across the micron-scale core of the fiber (~ 5 ns), which is much smaller than the excited-state lifetime (240 ns) [16, 21]. To maximize TPA, naively one

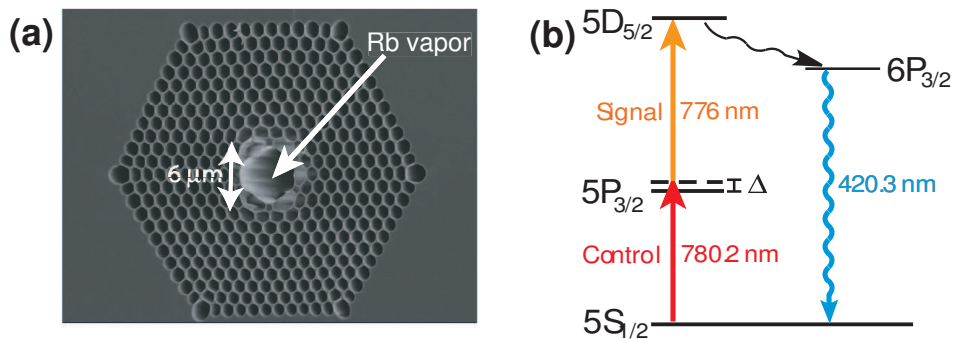


FIG. 1. (Color online) (a) Transmission electron microscope image of the cross section of the fiber (AIR-6-800) used in these experiments. Rubidium vapor is generated in the central core region, which then interacts with the light fields coupled into the core of the fiber. (b) TPA level scheme in ^{85}Rb used for performing all-optical modulation. An atom in the ground $5S_{1/2}$ state can simultaneously absorb a photon each from the 780-nm control and 776-nm signal beams to make a resonant transition to the excited $5D_{5/2}$ state. The signal photon can only be absorbed if the control photon is also present. A small fraction of the excited atoms decay through the $6P_{3/2}$ level emitting blue fluorescence.

might reduce the detuning from the intermediate level Δ as much as possible to resonantly enhance the interaction. However, since the 780-nm control beam experiences ordinary linear absorption in addition to TPA, the optimal detuning for maximal TPA is non-trivial. To theoretically model non-degenerate resonant TPA in a counter-propagating geometry, we numerically solve the following coupled nonlinear propagation equations for the intensities I_C and I_S of the control and signal beams respectively, taking into account the Gaussian velocity distribution of thermal atoms in the fiber,

$$\frac{dI_C}{dz} = -\alpha I_C - \beta I_C I_S, \quad (1)$$

$$\frac{dI_S}{dz} = \beta I_C I_S, \quad (2)$$

for $0 < z < L$, where $L \sim 1$ cm is the sample length, α is the linear absorption coefficient and β is the TPA coefficient. Our theoretical calculation shows that maximum TPA is attained at a detuning of approximately 600 MHz from line center, which is consistent with our experimental observation. For the Rb-PBGF system, using an OD ~ 100 which corresponds to an atomic density $N = 2 \times 10^{13}$ atoms/cm³ and a homogeneous decay rate $\gamma \sim 50$ MHz due to transit-time broadening [16], we estimate an effective TPA coefficient $\beta = 50$ cm/W. We also conclude that the ratio β/α plays a critical role in determining the effective TPA in this system. The amount of TPA for a specified amount of optical power is found to first increase with OD but then saturate for OD $\gtrsim 100$; increasing the atomic number further has no effect. We confirm this by measuring TPA at different optical depths and observing that it is nearly independent of OD, which is consistent with the theoretical prediction since β/α is independent of atomic density.

The experimental setup is shown in Fig. 2. Hollow-core PBGFs (Crystal Fiber AIR-6-800) are mounted inside a vacuum chamber with a Rb source attached. The fiber has a 6- μm diameter hollow core and guides light in the 750-810 nm wavelength range with a fundamental mode area given by 10^{-7} cm². The signal and control beams (at 776 nm and 780.2 nm) have identical circular polarizations and are coupled counter-propagating into the core of the fiber to eliminate Doppler broadening. The 776-nm laser is scanned across the TPA resonances while the 780-nm laser is tuned near the D_2 line of ^{85}Rb . A 3-mW highly non-resonant vapor generation beam at 805 nm with orthogonal polarization to the pump beams is also coupled into the fiber to evaporate Rb nanoclusters adsorbed on the inside walls of the core [25], and to generate a high OD ~ 100 . The OD generated is continuously monitored during the experiment using another laser at 795 nm scanning across the Rb D_1 line. A pick-off is used to directly measure the absorption from the 776-nm pump beam after it exits the fiber using a sensitive photodetector.

Figure 3 shows the transmission of the signal beam as it was scanned across the $5P_{3/2} \rightarrow 5D_{5/2}$ line of ^{85}Rb . The control beam was tuned to a frequency 600 MHz to the blue of the $5S_{1/2} \rightarrow 5P_{3/2}$ ($F = 3 \rightarrow F' = 4$) transition. We observed $>20\%$ absorption with less than 2 nW of total power in the fiber. The powers in the signal and control beams were 600 pW and 825 pW, respectively. A transit-time limited transmission profile is of the form $T(\omega) = 1 - Ae^{-\tau|\omega|}$, where ω is the detuning from two-photon resonance and τ is the transit-time [26]. We fit the data in Fig. 3 to a sum of five exponential functions corresponding to the hyperfine states ($F' = 1, 2, 3, 4, 5$) of the excited $5D_{5/2}$ level in ^{85}Rb and extract the transit-time to be 5.3 ns. This agrees very well with our previous measurement of the transit-time in a Rb-PBGF system [16, 21]. We also extract the TPA cross-

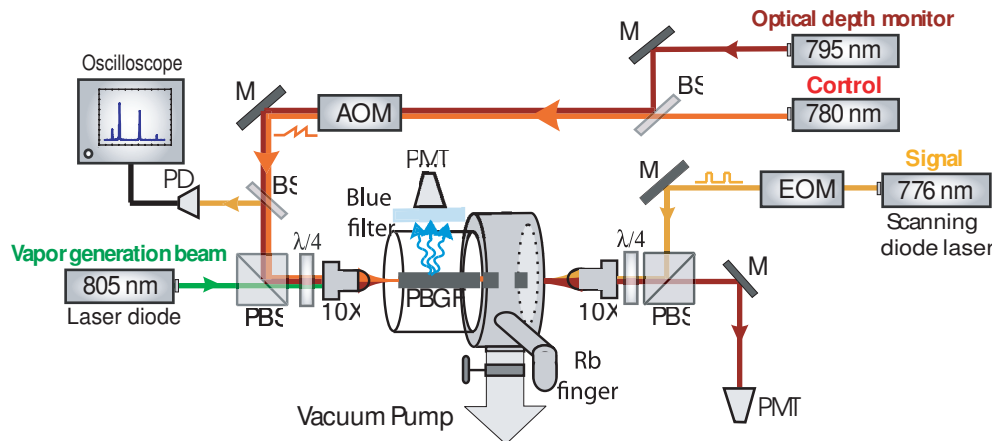


FIG. 2. (Color online) The polarizations of the signal and control beams are made identical (circular) using polarization beam splitter (PBS) cubes and quarter ($\lambda/4$) waveplates, and the beams are then coupled counter-propagating into the fiber. A pick-off is used to monitor the transmission of the signal beam using a sensitive photodiode (PD). A strong (3 mW) off-resonant vapor generation beam is also coupled into the fiber to generate the desired atomic density and optical depth. The optical depth is monitored during the experiment using a weak 795-nm beam scanning across the D_1 resonance of Rb. An electro-optic modulator (EOM) driven by a function/waveform generator is used to make square pulses from the signal beam for the pulsed measurement shown in Fig. 4. An acousto-optic modulator (AOM) driven by another function generator is used to ramp the power of the control beam as a sawtooth wave for the measurement shown in Fig. 5.

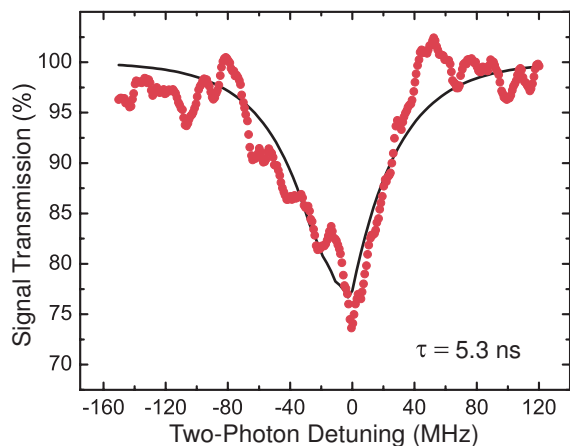


FIG. 3. (Color online) Transmission (red dots) of the signal beam as it is scanned across two-photon resonance showing $>20\%$ absorption with 1.4 nW of total power in the fiber. The OD attained in the fiber is ~ 200 . The solid black line shows a fit produced by summing five exponential lineshapes corresponding to the accessible hyperfine states of $5D_{5/2}$ level, from which the transit time τ was determined to be 5.3 ns.

section $\sigma^{(2)}$ to be $5 \times 10^{-12} \text{ cm}^4/\text{W}$, which agrees with the theoretically calculated value taking into account the transit time of the Rb atoms as the dominant decay rate and averaging over the Doppler profile.

In order to confirm that the response time of our system is comparable to the transit time, we set up a pulsed measurement. An electro-optic modulator (EOM) is used to amplitude modulate the signal beam [see Fig. 2] and make square pulses of varying duration (from 5 - 200 ns), and the TPA is measured for each pulse width. The pow-

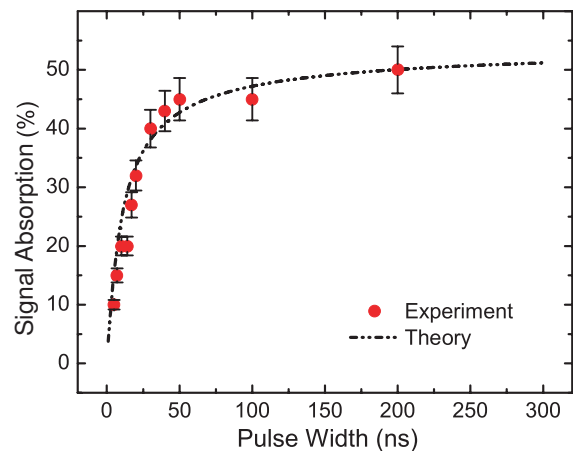


FIG. 4. (Color online) Absorption (red dots) of the pulsed signal beam at two-photon resonance versus pulse width. Error bars indicate measurement noise (one standard deviation). The experimental data agree very well with the theoretically predicted curve (dotted black line) for a transit time $\tau = 5$ ns of the Rb atoms across the fiber core and corroborates that the response time of the system is determined by the transit time.

ers in the two beams were kept 5-10 times higher than in the previous CW measurement to improve the signal-to-noise ratio. The TPA is found to reduce with decreasing pulse width, with a profile as shown in Fig. 4. The experimental data agree very well with the theoretically predicted curve for a transit time $\tau = 5$ ns. As there are no fitting parameters in the theory curve, the pulsed measurement concurs with the CW linewidth measurement to confirm the response time of our system as 5

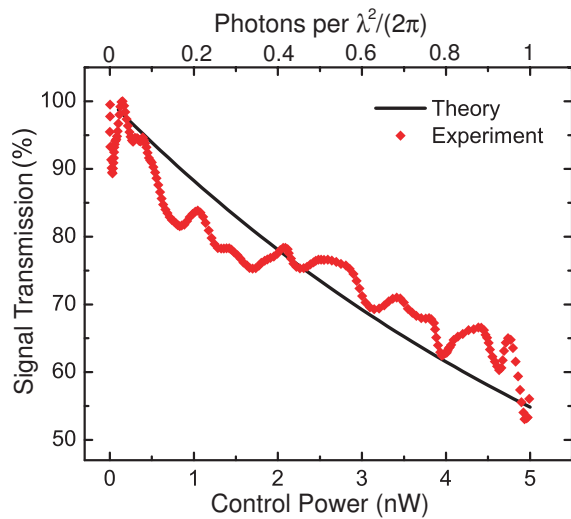


FIG. 5. (Color online) Transmission (red dots) of the signal beam at two-photon resonance with varying power in the control beam. Up to ~ 3 dB attenuation in the signal beam is observed with less than 5-nW of control power which corresponds to an energy density less than one photon per $(\lambda^2/2\pi)$. The solid black line shows the theoretical prediction from numerical simulation of the nonlinear propagation equations.

ns. Modulation bandwidths up to 50 MHz are thus possible even with this sensitive scheme. Furthermore, this implies on average only 12 and 16 photons in the signal and control beams, respectively, are interacting with the atoms within the transit time, for the 25% modulation shown in Fig. 3. 16 photons at the 780-nm wavelength correspond to 5 aJ of energy.

Finally, we measured the attenuation induced on the CW signal beam with varying power of the control beam at two-photon resonance. An acousto-optic modulator (AOM) is used to amplitude modulate the control beam [see Fig. 2] and ramp its power as a sawtooth wave. Figure 5 shows the data for one continuous ramp. We observe larger attenuation in the signal beam with higher control beam power, and up to 3 dB attenuation is achieved with less than 5-nW power, which corresponds to an energy density of less than one photon per $(\lambda^2/2\pi)$. The experimental data match the theoretical trend reasonably well.

In summary, we demonstrate few-photon all-optical modulation using non-degenerate TPA in thermal Rb atoms confined to a hollow core PBGF. We show 20% attenuation of a signal beam with a switching energy of only 5 attojoules, corresponding to about 16 photons. The switching energy density is less than one photon per atomic cross-section. We also characterize the time response of the system to be 5 ns, which enables large modulation bandwidths up to 50 MHz for a highly sensitive atomic vapour-based scheme. Our results compare favourably with those achieved in previous experiments, including more elaborate and complicated cold atom se-

tups [2–5]. Moreover, our system enables us to explore a wide space of atomic density and optical intensity in a controllable manner and holds promise for integration with fiber-optic communication networks [27]. These results show the potential of a Rb-PBGF system for exploring quantum nonlinear effects at ultralow powers.

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