Generation of large alkali vapor densities inside bare hollow-core photonic band-gap fibers

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Abstract: We demonstrate the ability to generate extremely large rubidium densities in uncoated hollow-core photonic band-gap fibers using light-induced atomic desorption. Once the fiber is exposed to Rb vapor for 1-2 weeks, and this atomic source is removed, the fiber yields large desorbable densities for an extended period of time. We show that optical depths greater than e^{-1200} can be created within seconds. Our observed Rb densities are several orders of magnitude larger than any previously reported to be generated optically, and allow for the demonstration of a relatively easy-to-use fiber-based vapor cell capable of producing large optical depths without the need for thermal tuning.

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1. Introduction

The field of quantum optics is rapidly advancing towards a regime in which optical nonlinearities can be induced with single photons [1]. In recent years, two general approaches have been implemented to achieve ultralow-level nonlinear interactions: 1) Cavity-based geometries [2], and 2) waveguide-based geometries where light is tightly confined and can interact with optically-dense atom ensembles on length scales that far exceed that which can be achieved with free-space focusing [3,4].

Hollow-core photonic bandgap fibers (PBGF) are a relatively new addition to the optical fiber family, wherein an air-core is surrounded by a Bragg-like hexagonal lattice of glass and air [5]. Unlike traditional step-index fibers in which light is guided by total internal reflection, in PBGF light is guided in the hollow-core by diffraction. Thus, PBGFs are attractive for studying light-atom interactions with all the advantages of waveguide geometries [3,6].

Alkali atoms such as Rb and Cs are well suited for photonic applications such as EIT-based switching [7], pulse storage/retrieval [8], atomic clocks and frequency standards [9], and laser frequency stabilization [10]. For some of these applications, such as EIT-based switching and light storage and retrieval, maximizing the optical density proves to be a critical requirement [11]. Thus, there is a vigorous effort at present to fill hollow waveguides with optically dense alkali vapors. However, despite widespread demonstration of light-matter interactions with molecular and atomic gases confined in hollow-core PBGF [3,12-15] and semiconductor waveguides [16], there have been only a few reports of photonic interactions with confined alkali vapors [4,17,18]. Alkali vapors are problematic since the atoms adsorb to the core walls, and a thermal equilibrium vapor cannot be established until all surfaces have "ripened" [19]. The extreme aspect ratio of an optical fiber suggests that the core walls will ripen on timescales of a year.

One way to obtain an appreciable Rb vapor in confined geometries is by light-induced atomic desorption (LIAD) [17,19,20]. LIAD is a process in which the ejection of sizable atomic densities from an alkali-exposed glass surface is induced by sudden illumination of a relatively weak beam. The mechanism by which LIAD works is still an open question, but it does not appear to be thermal [19,20]. Traditionally, the glass is pre-coated with a layer of long hydrocarbon chains such as siloxanes or paraffins [19,21]. We have previously described

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the coating of a hollow-core fiber with a monolayer of hydrocarbon moieties by microfluidic hydrolysis and condensation reaction, and we demonstrated the use of LIAD in such a fiber for generating large Rb densities [17]. Very recently, it has been reported [22] that alkali-atom desorption is possible from uncoated porous glass surfaces and that it may even be larger than that from hydrocarbon-coated glass.

Here, we present the first report of the large Rb densities that can be generated inside of an unmodified hollow-core PBGF by LIAD. We estimate on-resonance transmission as low as e^{-1200}, which represents the generation of optical densities that are several orders of magnitude larger than have ever been reported for light-induced desorption [23]. The ability to use unmodified PBGF without the need for further chemical processing considerably simplifies this technology. Furthermore, we have found that such uncoated fibers show higher transmission properties and better-defined core modes than coated fibers, which allows for photonic interactions at even lower light powers.

2. Experimental

We assemble the PBGF Rb vapor cell by mounting a 4.5-cm-long, 6-μm core-diameter (Crystal Fiber Air-6-800) fiber inside a 4" diameter × 2" length steel cell with glass windows attached to an ultrahigh vacuum station. A source of molten Rb is attached to the sample cell, as shown in Fig. 1. Ambient rubidium vapor density is controlled by varying the temperature of the cell and the cold-finger assembly and is monitored by a resonant laser beam that passes through the cell without coupling into the fiber. The 795-nm probe beam is obtained from a diode laser and is scanned across the Rb D_1 transition (795 nm). This beam is coupled through the vacuum cell windows into the fiber core with a 10× microscope objective located outside of the vacuum assembly. The transmitted probe light is routed to a photomultiplier tube (PMT) for measurement. Part of the probe beam is sent to a CCD camera that images the front face of the fiber to assure coupling to the core mode. The desorption beam is obtained from a CW Ti:sapphire ring laser at 805 nm. This beam is coupled into the fiber in an analogous way to the probe beam, but in a counter-propagating geometry, to reduce background light on the PMT. Typical transmission through a fiber-cell system with a pristine (non-Rb-exposed) fiber is 12-35%, which includes losses at the objectives and at the vacuum cell windows and due to fiber coupling. The fiber bandgap extends from 750 nm to 810 nm, and thus accommodates both the probe and desorption beam wavelengths. We typically use 10 nW of transmitted probe power and 20-90 mW of incident desorption beam power.

3. Results

We illustrate a significant difference between optical interactions with the ambient Rb density and with the Rb that is generated from—and is confined to—the fiber. Figure 2 shows the Doppler-broadened absorption profiles for the $F=2 \rightarrow F'=2,3$ transitions in the D_1 line of ^{85}Rb for each of these conditions. We desorb a small amount of Rb within the fiber to roughly match the optical depth seen in the ambient loading cell. The absorption lines fit well to Voigt profiles that account for both the homogeneous (Lorentzian) and inhomogeneously Doppler-broadened (Gaussian) lineshapes.

As shown in Fig. 2, the absorption profile of the fiber-borne Rb is considerably wider than that of the ambient Rb vapor. Fitting the in-cell Rb absorption lines to a Voigt profile that assumes a 5.75 MHz natural linewidth [24] yields a FWHM Doppler width of 570 MHz, corresponding to a cell temperature of 380 ± 30K. In this case, the natural linewidth is overshadowed by the Doppler-broadened linewidth. The confinement of hot Rb atoms to the 6 μm-diameter core introduces a significant contribution of transit-time broadening. From a conceptual perspective, the absorption of probe power can only take place on average during the time it takes for hot Rb atoms to traverse the light mode. Limiting light-matter interactions to this timescale effectively broadens the absorption lines, which is known as transit-time

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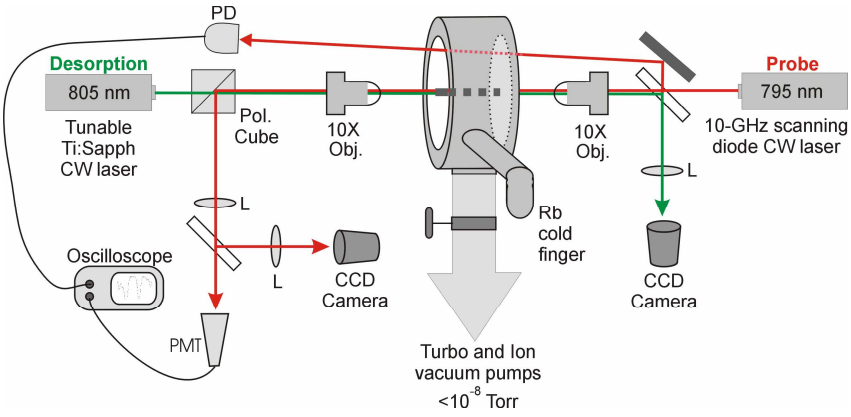

Fig. 1. A schematic of the experimental layout: A 4.5-cm-long hollow-core photonic bandgap fiber is mounted inside a steel chamber with glass windows connected to an ultrahigh vacuum system. 795-nm probe light and 805-nm atom-desorbing light are coupled into the fiber in counter-propagating geometry with 10× microscope objectives, and the coupling is monitored using a CCD camera. The probe beam can alternately be passed through the cell and detected by a photodiode to monitor the ambient cell Rb density. The coupled probe light is detected with photomultiplier tube (PMT). L: lens; PD: photodiode.

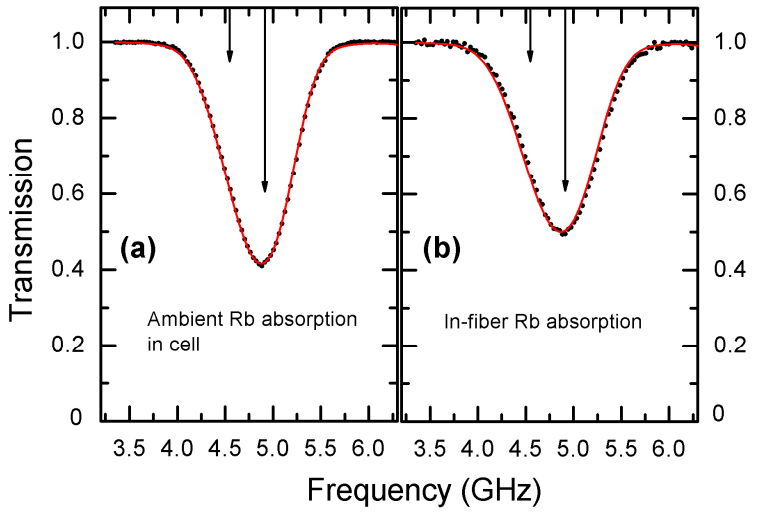

Fig. 2. Absorption profiles of F=2→F'=2,3 transitions in the D_1 line of ^{85}Rb. (a) The absorption profile of the ambient Rb vapor used to load the hollow-core fiber, as seen by passing light adjacent to the fiber. The position and relative oscillator strengths of the two transitions are indicated by the black arrows. The data is fit (black line) with a Voigt profile that assumes a natural linewidth of 5.75 MHz and yields a FWHM Doppler width of 575 MHz. (b) The through-fiber absorption profile in the presence of weak Rb desorption. The solid line is a fit to a Voigt profile with a Lorentzian component linewidth of 20 MHz, corresponding to transit-time broadening, and to a Doppler-broadening Gaussian component with a FWHM of 670 MHz. The key feature of note is that the through-fiber absorption profile is considerably broader than that of the ambient Rb vapor.

broadening. This phenomena is typically only observed in very precise measurements, such as in saturable-absorption spectroscopy or in two-photon absorption spectroscopy, in which for typical light-mode sizes and gas temperatures the broadening is on the order of kHz [25]. However, under the unique conditions and geometries of hollow-core optical waveguides, transit-time broadening is on the order of tens of MHz [26] and can significantly contribute to

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Doppler-broadened absorption lineshapes. Estimating the transit-time of a hot vapor traversing a Gaussian optical mode, the Lorentzian linewidth broadening is given by [27],

$$\Gamma_{tt} = \frac{0.58}{\pi}\frac{u}{r_{1/e}} = \frac{0.58}{\pi r_{1/e}}\sqrt{\frac{2k_B T}{m}}, \quad (1)$$

where u is the average velocity of the Rb atoms, $r_{1/e}$ is the light-mode radius, T is the temperature in Kelvin, k_B is the Boltzmann constant, and m is the atomic weight. Approximating $r_{1/e} \sim 3$ μm and $T \sim 500$ K yields a transit-time broadening of $\Gamma_{tt} \sim 20$ MHz. Since the Doppler width also scales as $T^{½}$, the two are correlated, and we can fit the in-fiber absorption profile to a Voigt function, where only temperature and the number of atoms are the free parameters. Doing so, we obtain a fit that yields a temperature of 520 ± 40 K for desorbed Rb atoms in the PBGF. Comparing this with our obtained temperature of 380 ± 30 K from fits to the ambient Rb absorption profiles from the front cell [Fig 2(a)], our data conclusively indicates that the Rb atoms desorbing from the glass core are hotter than the ambient temperature. Because of the transient nature of the Rb vapor density, this apparent temperature represents the kinetic energy of the Rb vapor, and does not necessarily reflect a thermally-equilibriated fiber temperature.

We have previously reported that transverse transit and dephasing collisions with the core walls is responsible for wide electromagnetically-induce transparency peaks in coated hollow-core fibers [17]. It should be noted, however, that neither Light *et al.* [18], nor Yang *et al.*[4] have reported any transit-time broadening in PBGF or ARROW waveguides, respectively. Thus, because the ambient Rb vapor and fiber-confined vapor behave differently, we examine the possibility of removing the ambient Rb density once the fiber has been "loading" for a few weeks, which guarantees that any Rb in the system is wholly fiber borne. This protocol has a further added operational advantage: The primary failure mechanism of our cells over the course of months to a year has been the cracking of the cell windows where it is continually exposed over a long period of time to a corrosive Rb vapor. Thus, the ability to operate such cells with only intermittent periods of ambient Rb-glass window interactions is expected to drastically increase the lifetime of fiber-based vapor cells.

We have studied the generation of Rb by LIAD in pre-loaded fiber. Rb is loaded into the fiber by exposing it to ambient Rb vapor at 70 ºC for 3 weeks. Once the fiber demonstrates strong light-induced desorption, the temperature is reduced to 20 ºC, thereby removing the ambient Rb vapor density by two orders of magnitude to about 2×10^{-6} Torr, and below our measurement resolution. Even in the absence of constant Rb-loading, we observe a strong LIAD signal. To the best of our knowledge, the amount of Rb generated in the fiber *in-vacuo* is on the same order as that generated in the presence of constant Rb-loading. We have used this fiber daily for weeks, over a wide range of desorption cycles. We have observed that while the total amount of available (desorbable) rubidium in a pre-loaded fiber decreases with use, the near instantaneous densities of vapor generated with a strong desorption pulse (~ 40-60 mW) remains nonetheless very large at least for a few weeks (on the order of fifty 1-20 s desorption events).

Figure 3 shows absorption profiles from Rb density generated in a pre-loaded fiber. The profiles of three different Rb densities are shown. It is difficult to estimate with certainty the optical depths at high Rb densities. At optical depths greater than 4, there is insufficient transmission on resonance to directly measure the extinction with a standard photodetector. A suitable technique to estimate optical depth from the absorption profile is to fit the transmission trace to a set of Voigt profiles, where most of the information regarding the vapor density lies in the wings and rising slopes between resonances. There are six natural Rb resonances within the investigated spectral range. We fit a linear superposition of integrated Voigt profiles of six of the Rb lines weighted by their abundance, oscillator strengths, and degeneracy, as shown in Fig. 3. The temperature of the Maxwell-Boltzmann distribution and

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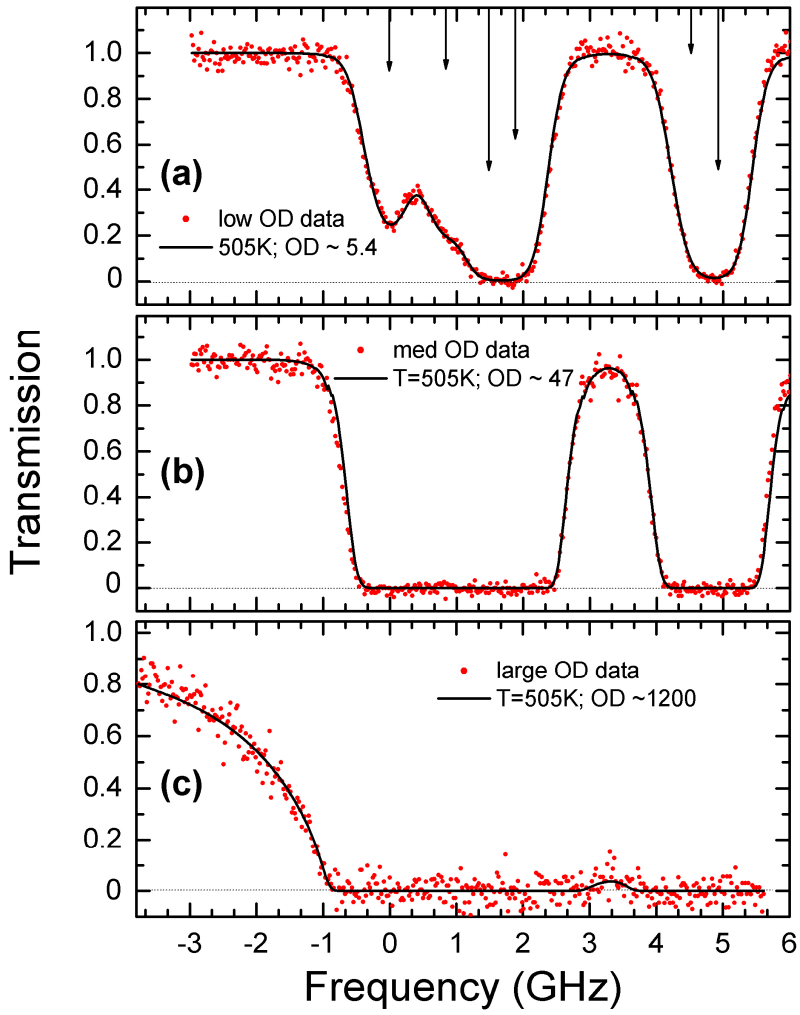

Fig. 3. Absorption profiles of lines in the D_1 transition of ^{87}Rb and ^{85}Rb, representing various densities generated by light-induced atomic desorption (LIAD) in a hollow-core optical fiber that has been previously exposed to Rb vapor. The frequency scale is zeroed on the lowest energy D_1 transition (^{87}Rb F=2→F'=2). The top panel shows a relatively low optical depth of Rb. The black line is a fit to six Voigt lines that include the effects of transit-time broadening (see text). The position and relative absorption strength (accounting for natural abundance and oscillator strength) [24] are represented by the black arrows and their lengths, respectively. The middle panel shows a moderately high optical depth generated by LIAD. The black line is a free fit to the data with the same expression as that in the top panel. The bottom panel shows the absorption profile of a large density, with a conservatively estimated optical depth of ~1200 between the F=3→F'=2,3 resonances of ^{85}Rb (see text).

absorption pre-factor are freely varying parameters that are used to optimize the fit to the observed data. By comparing the absorption pre-factor to a standard pre-factor that generates an optical depth of 1 at the highest absorption point (between ^{85}Rb F=2→F'=2,3), we deduce the optical depth as seen through the fiber. A temperature-dependent transit-time broadening (see Eq. 1) is also included in the Lorentzian linewidth as part of the Voigt profile.

At lower optical depths, as shown in Figs. 3(a) and 3(b), the fits yield a temperature of 505±30K. Depending on the timing and length of the desorption pulse, a wide range of optical depths can be produced. Figure 3(b) shows an absorption profile for a moderate Rb density. Here, the maximum optical depth (OD) is defined as,

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$$OD = -\ln T(\nu)_{\min} \ , \tag{2}$$

where $T(\nu)_{\min} \sim e^{-47}$ is the minimum transmission. When the intense desorption beam is first turned on, a large Rb density is generated within a few milliseconds. As seen in Fig. 3(c), sufficient Rb density is created to give near complete absorption across the entire D_1 line.

Estimating the optical depth of the largest Rb densities we can generate by LIAD is complicated by the fact that at such a large atomic density and desorption beam intensity, additional broadening mechanisms may be present, which further complicates the absorption lineshapes. The best indication of the optical depth lies in the fact that the region between the ^{85}Rb F=3→F'=3 and ^{85}Rb F=2→F'=2 lines (at a detuning of 3.3 GHz) experiences almost complete absorption. In an attempt to estimate the optical depth implied by the absorption presented in Fig. 3(c), we use the fitting parameters obtained from fits to the low-to-moderate OD absorption data, and further assume a ~5% transmission peak at 3.3 GHz. We thus estimate that the optical depth of the absorption profile shown in Fig. 3(c) to be approximately 1200. It is possible for such rapid generation of Rb density at the onset of desorption that the atoms are considerably hotter than 500 K. While we have no indication that this occurs, in such a case the OD would be significantly lower than our estimates.

A comparison between a hollow-core fiber based vapor cell and a standard glass vapor cell demonstrates several key advantages to a fiber-based device: The primary advantage lies in an ability to dynamically and rapidly modulate the optical depth. Our demonstration of the ability to generate ODs between 1000 and 0 within a few seconds is unique. Furthermore, because the desired Rb densities in standard vapor cells are obtained thermally, the absorption profile is ever-changing due to increasing Doppler widths. Thus it is impossible to dynamically change the optical depth in a standard cell without changing the spectral profile. As the data in Figs. 3(a) and 3(b) shows, we can achieve large increases in optical depth without corresponding temperature changes. Thus, this type of vapor cell may find application as an isotopically-pure filter cell [28] where, for example, the fiber is loaded with pure ^{85}Rb and buffer gas, and a sufficiently high Rb density is generated by LIAD such that a large optical depth is obtained 1.6 GHz from the line centre, but no absorption is present at 1.9 GHz. This would act to completely filter out one of the ^{87}Rb D_1 doublets (F=2→F'=1,2), while passing the other (F=1→F'=1,2).

Another major advantage of our cell geometry is an expected abatement of radiation trapping effects [29]. Radiation trapping occurs in optically dense media when spontaneous fluorescence is absorbed by ground-state atoms before it can escape (or be detected). This effect is exacerbated in media with a large transverse-to-axial density ratio. Our geometry, wherein the aspect ratio of transverse-to-axial lengths is >5000 and where the interaction region (light mode area) essentially fills the entire transverse extent of the vapor is ideal for minimizing radiation trapping. Thus, the hollow-core PBGF vapor cell may be uniquely suited for quantum nonlinear optical experiments where dynamically varying and large optical depths are needed, but where radiation trapping is a concern.

4. Conclusion

We have demonstrated strong light-induced atomic desorption of Rb atoms in uncoated, hollow-core PBGFs. Previously, LIAD was thought to be possible only from fibers that have been microfluidically-treated with hydrocarbons [17]. Thus, the discovery that desorption in a bare fiber can generate at least as much Rb as has been seen in a coated fiber, is significant, since it considerably simplifies their use. We show that the absorption profiles of the ambient cell Rb and fiber-borne Rb densities to be distinct, demonstrating that the in-fiber absorption profile is wider due to transit-time broadening and due to atoms desorbing at temperatures higher than the walls. Subsequently, we have found that once the fiber is exposed to Rb vapor at 50-100 ºC for a few weeks, the ambient rubidium may be removed and the fiber can be repeatedly used for an extended period of time *in-vacuo*. Such a fiber can generate large optical depths, estimated to exceed 1000 on the ^{85}Rb D_1 resonance. With improved design and miniaturization, we foresee the ability to manufacture small and self-contained fiber-based

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vapor cells in the near future. We motivate the future use of such a fiber-based vapor cell for optical filtering of particular hyperfine levels and for sensitive quantum nonlinear optical measurements where large optical depths are needed and where radiation trapping must be minimized.

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