In recent years, Rydberg atoms have emerged as leading candidates for neutral atom based quantum information processing [1–4] and quantum simulations [5–7]. The long-lived quantum states and the precisely tunable dipolar interaction, leading to blockade [8], can be used to prepare a mesoscopic atomic ensemble exhibiting quantum correlations and entanglement [9]. Such systems have already been used to demonstrate a quantum phase gate [10] and simulator [7] in free-space. Interfacing interacting Rydberg atoms with microfabricated devices is a very attractive choice for building compact and scalable hybrid quantum devices. Coherent Rydberg excitation has been reported for an atom chip [11], a µm-sized vapor cell [12], a hollow-core photonic crystal fiber [13, 14] and has been proposed for a superconducting resonator [15], with each platform having its own advantages and disadvantages.

Here, we present an alternative, but highly viable, platform for atom-based quantum networks by interfacing cold Rydberg atoms with a single-mode optical nanofiber (ONF). To date, ground state cold atom-ONF hybrid systems [16, 17] have shown tremendous potential for a new generation of quantum devices. The small cross-section of the evanescent field from an ONF, as a result of the exponential radial decay from the fiber waist, leads to a large co-operativity [13, 19], the key to many quantum optics experiments. The high intensity and field gradient can be used to optically trap atoms in a one-dimensional array [20, 21], thereby enabling the study of one dimensional, many-body physics, or can be exploited for quadrupole transition enhancement [22]. Cooperative effects, such as the generation of a collective entangled state [23], have been demonstrated in such a system using ground-state, neutral Cs atoms. In addition, atoms in the evanescent field region are intrinsically coupled to an optical bus in the form of the fiber-guided mode. This can lead to low-loss transfer of information to and from the interaction region [24], a prerequisite for Rydberg-based quantum repeaters in fiber-coupled cavities [25].

Aside from endeavours to combine ONFs and ground state, neutral atoms, work on Rydberg excitation next to ONFs has, to-date, been limited to theoretical proposals due to the difficulty in generating highly excited atom states within a few 100 nm of surfaces, e.g. dielectrics or metals, and the problem of induced electric fields - even at distances as large as ~100 µm - by adsorption of atoms on the surface [26, 27]. In this Letter, we report on evanescent field assisted Rydberg excitation at submicron distances from the surface of an ONF, which is embedded in a 87Rb atomic ensemble in a magneto-optical trap (MOT). A ladder-type, two-photon excitation scheme [28] is used to excite the atoms to a Rydberg state and a trap loss method [29] is used to probe the Rydberg excitation. We implement a rate equation model [30] to determine the rate of population transfer to the Rydberg state. Both coherent two-photon excitation and incoherent two-step excitation is demonstrated. A density matrix based model is developed for the three-level ladder-type system. The experimental results are consistent with the developed model.

The experiment consists of an ONF, with a waist diameter of ~400 nm that is single-mode for 780 nm, embedded in a cold atomic ensemble of 87Rb. A schematic of the experimental setup is given in Fig. 1(a). The ONF is fabricated by exponentially tapering a section of SM800-125 fiber (cut-off wavelength 697 nm) using a H:O flame-brushing technique [31]. To guarantee a low-loss ONF in the ultrahigh vacuum (UHV) chamber, we ensure that the tapering process is adiabatic and that the fiber itself is very clean. During the experiment, 100 µW of 1064 nm light is passed through the ONF from each side to keep the fiber hot and avoid atom deposition on the ONF [32]. The 87Rb atoms are cooled to ~120 µK using a standard MOT configuration of three pairs of retro-reflected
cooling and repump beams. The 780 nm cooling beam is 2π×14 MHz red-detuned from the 5S_{1/2}(F = 2) → 5P_{3/2}(F' = 3) transition and the repump is on resonance with the 5S_{1/2}(F = 1) → 5P_{3/2}(F' = 2) transition. The total powers in the cooling and repump beams are 42 mW and 2 mW, respectively. Using a magnetic field gradient of ~24 G/cm at the center of the MOT, typically 10^7 atoms are trapped, and the typical Gaussian FWHM of the MOT is 0.5 mm. The free-space MOT fluorescence is collected by an achromatic doublet (f = 150 mm) and is divided into two parts using a 50:50 beam-splitter. One half of the signal is collected by a PMT to measure the instantaneous number of atoms in the MOT. The other half is imaged by an EMCCD camera to obtain the atom cloud density profile. The MOT fluorescence that is coupled to the guided-mode of the ONF is separated from the light of other wavelengths using an assembly of dichroic mirrors and bandpass filters, before being delivered to an SPCM. The overlap between the cold atom cloud and the ONF is optimized by maximizing the photon count at the SPCM using three pairs of Helmholtz coils. With an optimized overlap, the SPCM count is proportional to the atom density, and, hence, the PMT signal, for a given Gaussian full-width-half-maximum (FWHM).

The Rydberg excitation is driven by a ladder-type, two-photon process, shown in Fig. 1(b). The 780 nm light is provided by the cooling beams, while the 482 nm light is derived from a Toptica TA SHG Pro system and coupled into the nanofiber. The frequency of the 482 nm laser is stabilized to a vapor cell electromagnetically induced transparency (EIT) signal [28] and frequency shifted using an electro-optic modulator. The details of the locking scheme are given in the Supplemental Material [33]. The 482 nm light can be switched on and off using a combination of an AOM and a mechanical shutter to ensure complete cut-off. This light is coupled to the ONF using a pair of dichroic mirrors (DMLP650) and interacts with the atoms in the MOT via the evanescent field. The output light power is measured using a power meter to provide an estimate of the 482 nm power at the nanofiber waist; typical output values of about 16 μW recorded. Note that, at 482 nm, the ONF supports the fundamental mode, HE_{11}, as well as the TE_{01}, TM_{01}, and HE_{21} higher order modes.

The experimental sequence is simple. First, the MOT is loaded to saturation for 8 seconds. The normalized number of atoms in the MOT at any time, t, can be obtained from the PMT signal and is expressed as

$$N(t) = \frac{N_{\text{tot}}(t)}{N_0} = (1 - e^{-\gamma_m t}),$$

where $N_{\text{tot}}(t)$ is the total number of atoms in the MOT at time $t$, and $N_0 = L/\gamma_m$ is the steady-state number of atoms in the MOT. $L$ is the loading rate of atoms into the MOT from the background vapor and $\gamma_m$ is the loss rate of atoms from the MOT. A typical loading curve is shown in Fig. 1(c). Fitting the PMT signal with Eq. 1 gives $\gamma_m$, the atom loss rate from the MOT. When the MOT is loaded to saturation, the 482 nm laser propagating in the ONF is turned on. Only those atoms in the evanescent field of the nanofiber can interact with both the 780 nm and 480 nm light and, therefore, participate in the two-photon Rydberg excitation. The newly formed Rydberg atoms leave the cooling cycle and escape the MOT - this introduces a new loss rate, $\gamma_R$, which includes any other loss processes, such as ionization of atoms post Rydberg excitation. The new loss mechanism starts bleeding the MOT of atoms and a new equilibrium is established over time, determined by the total loss rate, $\gamma = \gamma_m + \gamma_R$. The time-dependent decay of the MOT can be written as

$$N(t) = 1 - \frac{\gamma_R}{\gamma_k} (1 - e^{-\gamma_k(t-t_0)}),$$

where $t_0$ is the time at which the 482 nm laser is switched on.

The time evolution of the atom number in the MOT is fitted with Eq. 2 to obtain $\gamma_R$. Assuming all atoms excited to the Rydberg state are lost from the cooling cycle, $N_0\gamma_R$ is the rate of formation of the Rydberg atoms.
excitations can be found in Ref. [35].

A detailed explanation of a similar mechanism affecting the ratio between coherent and incoherent processes is presented in this Letter. A detailed explanation of a similar mechanism affecting the ratio between coherent and incoherent excitations can be found in Ref. [35].

Figure 2 shows the variation of $\gamma_R$ as a function of $\Delta$, for three different values of the cooling laser detuning, $\delta$. The position of the coherent peak, P1, changes with $\delta$ to satisfy the two-photon resonance condition. As we expect, the position of P2 does not change for $\delta = 10$ MHz and 14 MHz; however, there is a noticeable shift when $\delta = 18$ MHz. A detailed theoretical model would be needed to determine the reason for this observed shift. Finally, we investigated the dependence of $\gamma_R$ on the Rabi frequency of the $5P_{3/2} \rightarrow 5D_{3/2}$ transition for both the coherent and incoherent processes. To perform this experiment, $\delta$ was set to 14 MHz and $\Delta$ was set to the maximum of either P1 or P2. The power of the 482 nm laser was then varied and $\gamma_R$ was measured for both peaks and both transitions. The results are shown in Fig. 3. We compare the dependence of the observed decay rates on the frequencies and intensities of the driving optical fields by considering a three-level density matrix model for a population of atoms driven by two coherent optical fields of constant intensity. The basic model includes a mixing rate, $\Lambda$, at which atoms enter and leave the evanescent field of the 482 nm light, coherence dephasing due to the thermal motion of atoms, and mutual incoherence of the optical fields. It ignores other aspects of the experimental geometry. The cooling field Rabi frequency, $\Omega_p$, is fitted from the splitting of P1 and P2, and the 482 nm Rabi frequency, $\Omega_r$, is fitted to the P1 and P2 height data. The decoherence rates are fitted to the widths of P1 and P2. The full model is included in the Supplemental Material [33].

The mixing rate, $\Lambda$, is an important parameter for describing this experiment. The introduction of atoms in a mixed state to the interaction region boosts the incoherent production of Rydberg atoms well beyond that expected in a coherently driven system. The mixing rate may be changed experimentally by changing the cooling...
field detuning and thus the temperature of the atoms. The effect of this can be seen in Fig. 3 where a closer detuned trap with hotter atoms has a higher mixing rate and a larger incoherent peak than a further detuned, cooler trap.

The population of Rydberg atoms in the model, $\sigma_{rr}$, is related to the experimental decay rate, $\gamma_R$, by estimating the proportion of atoms in the MOT that are also in the fundamental mode of 482 nm light is 125 nm. The mixing rate, $\Omega$, is set to 0.6 MHz to fit the model to the experimental data in Fig. 4 and this value also agrees with other experimental observations. It corresponds to an interaction region extending about 100 - 200 nm from the fiber surface, noting that the 1/e decay length of the evanescent field of the fundamental mode of 482 nm light is 125 nm. The mixing rate, $\Omega$, is set to 0.6 MHz to fit the ratio of the incoherent to coherent peak heights in Fig. 4. This value is also consistent with the average flight time of atoms at 120 $\mu$K through an interaction region with a diameter, $d = 200$ nm, where $A \approx \bar{v}/d$, with $\bar{v}$ the average speed of the atoms.

The experimental decay rate clearly follows the theoretical curve at low $\gamma_R$, but diverges from the model above rates of 5 Hz. This may be due to production of Rydberg atoms beginning to saturate as $\sigma_{rr} \rightarrow 1$. This is not observed in the model, with a Rydberg population at P1 of $\sigma_{rr} = 0.033$ for $\Omega = 1.25$ MHz, which is well below saturation. The model only includes one cooling field, with a Rabi frequency equal to that of one cooling beam. $\Omega$, is assumed to be constant, whereas, in the experiment, atoms are subject to a time-varying interaction as they travel through the evanescent field. Averaging over these trajectories may give a more accurate relationship between the 482 nm intensity and the effective $\Omega_r$. The model also assumes that all atoms leaving the interaction region while in the Rydberg state are lost from the MOT. However, considering the lifetime of the Rydberg states and the temperature of the atoms, a significant proportion of these atoms could actually be recaptured directly into the MOT if they decay into the cooling cycle. Experimental determination of the saturation rates under different conditions and for different $n$ could allow any processes interfering with recapture, such as ionization, to be observed.

In this work we have reported on the generation of $n = 29$ level Rydberg atoms in a $^{87}$Rb cold atom ensemble surrounding an optical nanofiber and have shown that this is a very viable system for hybrid atom-nanofiber devices. Excitation of the Rydberg atoms is mediated via the optical nanofiber and they are estimated to be generated at no more than a couple of 100 nm from the surface where overlap between the two excitation fields (780 nm and 482 nm) is maximum. This is a significant advance on previous Rydberg generation close to dielectric surfaces [28] and opens up many avenues of research such as all-fibered quantum networks using Rydberg atoms, excited atom-surface interactions at sub-micron distances, including van der Waal’s interactions, effects on the Rydberg blockade or facilitation phenomena [30] in this new regime, stray electric field effects, for example, from the dielectric nanofiber, on energy levels and lifetimes of the atom, and, perhaps even more intriguing, the limitation of the maximum excited state ($n$ value) that can be generated close to the nanofiber due to the atom size increasing quadratically with $n$.

The versatility of this atom-nanofiber hybrid system could be extended to explore three-step Rydberg excitations [27] where the fiber would be single-mode for the light used to drive the atomic transitions. Hence, the mode overlap in the evanescent field would be increased and Rydberg generation efficiency should be improved. A loss in detection of the 420 nm light in the 6P $\rightarrow$ 5S decay channel could also provide an alternative, non-destructive mechanism for Rydberg atom detection. In addition, a comprehensive study of the coherent interactions in MOT-embedded nanofibers could extend this experimental technique beyond a qualitative confirmation towards an investigation of the behavior of Rydberg or other exotic states, e.g. Rydberg polarons [38], adjacent to optical nanofibers. Future work will focus on trapping atoms at defined distances from the nanofiber to explore limitations on $n$ and on a study of the influence of the nanofiber on Rydberg levels using EIT signals [28].

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conducted the experiments. J.E. did the theoretical modelling. S.N.C. conceptualized the work. All authors wrote the manuscript.

EXPERIMENTAL SETUP AND DETAILS FOR RYDBERG VAPOR CELL EIT

FIG. 5. Setup for vapor cell Rydberg EIT. BPD: Balanced photodiode, EOM: Electro-optic modulator.

A standard vapor cell EIT [39], as shown in Fig. 5, is used to lock the frequency of the 482 nm laser, although a modification is implemented to shift its frequency of the 482 nm. First, the 780 nm probe laser is locked to the $5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 2, 3)_{co}$ transition of $^{85}$Rb. An EOM is used to generate sidebands at 1.06632 GHz, one of which is resonant with the $^{87}$Rb $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transition. In the vapor cell, which is enriched with $^{87}$Rb, only the resonant sideband participates in the EIT process and the resultant EIT peak is used to lock the frequency of the 482 nm laser. The frequency of the 482 nm laser can now be changed easily just by changing the frequency of the sideband.

THEORETICAL MODEL OF INTERACTION REGION

We use the Maxwell-Bloch equations for thermal atoms in a small interaction volume. We define the Rabi frequencies, $\Omega_p$ and $\Omega_r$, and the detunings from resonance, $\Delta_p$ and $\Delta_r$.

$$\partial_t \sigma_{ss} = \left[ i \frac{\Omega_p}{2} \sigma_{ps} + C.C. \right] + \Gamma_p \sigma_{pp} + \Gamma_r \sigma_{rr}$$

$$- A(\sigma_{ss} - \frac{1}{2}),$$

$$\partial_t \sigma_{pp} = \left[ i \left( \Omega_r \sigma_{rp} - \frac{\Omega_p}{2} \sigma_{ps} \right) + C.C. \right] - \Gamma_p \sigma_{pp}$$

$$- A(\sigma_{pp} - \frac{1}{2}),$$

$$\partial_t \sigma_{rr} = \left[ i \Omega_r \sigma_{rp} + C.C. \right] - (\Gamma_r + A) \sigma_{rr},$$

$$\partial_t \sigma_{ps} = - i \Delta_p \sigma_{ps} + i \frac{\Omega_p}{2} (\sigma_{s} s - \sigma_{p} p) + i \frac{\Omega_r}{2} \sigma_{rs}$$

$$- (\Gamma_p + A) \sigma_{ps},$$

$$\partial_t \sigma_{rp} = - i \Delta_r \sigma_{rp} + i \frac{\Omega_r}{2} (\sigma_{pp} - \sigma_{rr}) - i \frac{\Omega_r}{2} \sigma_{rs}$$

$$- (\Gamma_p + \Gamma_r + A) \sigma_{rp},$$

$$\partial_t \sigma_{rg} = - i (\Delta_p + \Delta_r) \sigma_{rg} + i \frac{\Omega_r}{2} \sigma_{ps} - i \frac{\Omega_p}{2} \sigma_{rp}$$

$$- (\Gamma_r + A + \gamma_0) \sigma_{rs}.$$
Bichler, A Rydberg quantum simulator, Nat. Phys. 6, 382 (2010)

[8] H. Labuhn, D. Barredo, S. Ravets, S. de Léséleuc, T. Macri, T. Lahaye, and A. Browaeys, Tunable two-dimensional arrays of single Rydberg atoms for realizing quantum Ising models, Nature 534, 667 (2016)

[9] H. Bernien, S. Schwartz, A. Keesling, H. Levine, A. Omran, H. Pichler, S. Choi, A. S. Zibrov, M. Endres, M. Greiner, V. Vuletić, and M. D. Lukin, Probing many-body dynamics on a 51-atom quantum simulator, Nature 551, 579 (2017)

[10] T. Wilk, A. Gaëtan, C. Evellin, J. Wolters, Y. Miroshnychenko, P. Grangier, and A. Browaeys, Entanglement of two individual neutral atoms using Rydberg blockade, Phys. Rev. Lett. 93, 063001 (2004)

[11] L. Isenhower, E. Urban, X. L. Zhang, A. T. Gill, T. Henage, T. A. Johnson, T. G. Walker, and M. Saffman, Demonstration of a neutral atom controlled-not quantum gate, Phys. Rev. Lett. 104, 010503 (2010)

[12] J. de Hond, R. van Bijnen, S. J. J. M. P. Kokkelmans, R. J. C. Spreeuw, H. B. v. L. van den Heuvel, and N. J. van Druten, From coherent collective excitation to Rydberg blockade on an atom chip, Phys. Rev. A 88, 062714 (2018)

[13] H. Kübler, J. P. Shaffer, T. Baluktsian, R. Löw, and T. Pfau, Coherent excitation of Rydberg atoms in micrometre-sized atomic vapour cells, Nat. Photon. 4, 112 (2010)

[14] D. Yu, L. C. Kwek, L. Amico, and R. Dumke, Superconducting qubit-resonator-atom hybrid system, Quantum Science and Technology 2, 035005 (2017)

[15] T. Nieddu, V. Gokhroo, and S. Nic Chormaic, Optical nanofibres and neutral atoms, J. Opt. 18, 053001 (2016)

[16] P. Solano, J. A. Grover, J. E. Hoffman, S. Ravets, F. K. Fatemi, L. A. Orozco, and S. L. Rolston, Chapter seven - optical nanofibers: A new platform for quantum optics (Academic Press, 2017) pp. 439 – 505.

[17] F. Le Kien, S. Gupta, V. Balýkin, and K. Hakuta, Spontaneous emission of a cesium atom near a nanofiber: Efficient coupling of light to guided modes, Phys. Rev. A 72, 032509 (2005)

[18] F. Le Kien, V. Balýkin, and K. Hakuta, Scattering of an evanescent light field by a single cesium atom near a nanofiber, Phys. Rev. A 73, 013819 (2006)

[19] P. Le Kien, V. Balýkin, and K. Hakuta, Atom trap and waveguide using a two-color evanescent light field around a subwavelength-diameter optical fiber, Phys. Rev. A 70, 063403 (2004)

[20] E. Vetsch, D. Reitz, G. Sagué, R. Schmidt, S. T. Dawkins, and A. Rauschenbeutel, Optical interface created by laser-cooled atoms trapped in the evanescent field surrounding an optical nanofiber, Phys. Rev. Lett. 104, 203603 (2010)

[21] F. Le Kien, T. Ray, T. Nieddu, T. Busch, and S. Nic Chormaic, Enhancement of the quadrupole interaction of an atom with the guided light of an ultrathin optical fiber, Phys. Rev. A 97, 013821 (2018)

[22] N. V. Corzo, J. Raskop, A. Chandra, A. S. Sheremet, B. Gouraud, and J. Laurat, Waveguide-coupled single collective excitation of atomic arrays, Nature 566, 359 (2019)

[23] J. A. Sedlacek, E. Kim, S. T. Rittenhouse, P. F. Week, H. R. Sadeghpour, and J. P. Shaffer, Electric field cancellation on quartz by Rb adsorbate-induced negative electron affinity, Phys. Rev. Lett. 116, 133201 (2016)

[24] A. Tauschinsky, R. M. T. Thijsse, S. Whiltlock, H. B. van Linden van den Heuvel, and R. J. C. Spreeuw, Spatially resolved excitation of Rydberg atoms and surface effects on an atom chip, Phys. Rev. A 81, 063411 (2010)

[25] A. K. Mohapatra, T. R. Jackson, and C. S. Adams, Coherent optical detection of highly excited Rydberg states using electromagnetically induced transparency, Phys. Rev. Lett. 98, 113003 (2007)

[26] J. O. Day, E. Brekke, and T. G. Walker, Dynamics of low-density ultracold Rydberg gases, Phys. Rev. A 77, 052712 (2008)

[27] S. Lee, K. Ravi, and S. A. Rangwala, Measurement of collisions between rubidium atoms and optically dense rubidium ions in trapped mixtures, Phys. Rev. A 87, 052701 (2013)

[28] J. M. Ward, A. Maimaiti, V. H. Le, and S. Nic Chormaic, Contributed review: Optical micro- and nanofiber pulling rig, Rev. Sci. Instrum. 85, 115101 (2014) https://doi.org/10.1063/1.4901098

[29] P. Anderson, S. Jalnapurkar, E. S. Moiseev, D. Chang, P. E. Barchya, A. Lezama, and A. I. Lvovsky, Optical nanoﬁber temperature monitoring via double heterodyne detection, AIP Advances 8, 055005 (2018)

[30] Supplemental material url to be inserted (2019).

[31] V. G. Minogin and S. Nic Chormaic, Manifestation of the van der Waals surface interaction in the spontaneous emission of atoms into an optical nanofiber, Laser Physics 20, 32 (2010)

[32] D. Grischkowsky, Coherent excitation, incoherent excitation, and adiabatic states, Phys. Rev. A 14, 802 (1976)

[33] M. Marcuzzi, J. Minár, D. Barredo, S. de Léséleuc, H. Labuhn, T. Lahaye, A. Browaeys, E. Levi, and I. Lesanovsky, Facilitation dynamics and localization phenomena in Rydberg lattice gases with position disorder, Phys. Rev. Lett. 118, 063606 (2017)

[34] D. P. Fahey and M. W. Noel, Excitation of Rydberg states in rubidium with near infrared diode lasers, Opt. Express 19, 17002 (2011)

[35] F. Camargo, R. Schmidt, J. D. Whalen, R. Ding, G. Woehl, S. Yoshida, J. Burgdörfer, F. B. Dunning, H. R. Sadeghpour, E. Demler, and T. C. Killian, Creation of Rydberg polarons in a Bose gas, Phys. Rev. Lett.
[39] R. P. Abel, A. K. Mohapatra, M. G. Bason, J. D. Pritchard, K. J. Weatherill, U. Raitzsch, and C. S. Adams, Laser frequency stabilization to excited state transitions using electromagnetically induced transparency in a cascade system, Appl. Phys. Lett. 94, 071107 (2009), https://doi.org/10.1063/1.3086305.