# **Important Notice to Authors**

No further publication processing will occur until we receive your response to this proof.

Attached is a PDF proof of your forthcoming article in *Physical Review Letters*. The article accession code is LQ17413.

Your paper will be in the following section of the journal: LETTERS - Atomic, Molecular, and Optical Physics

Please note that as part of the production process, APS converts all articles, regardless of their original source, into standardized XML that in turn is used to create the PDF and online versions of the article as well as to populate third-party systems such as Portico, Crossref, and Web of Science. We share our authors' high expectations for the fidelity of the conversion into XML and for the accuracy and appearance of the final, formatted PDF. This process works exceptionally well for the vast majority of articles; however, please check carefully all key elements of your PDF proof, particularly any equations or tables.

Figures submitted electronically as separate files containing color appear in color in the online journal. However, all figures will appear as grayscale images in the print journal unless the color figure charges have been paid in advance, in accordance with our policy for color in print (https://journals.aps.org/authors/color-figures-print).

# Specific Questions and Comments to Address for This Paper

The numbered items below correspond to numbers in the margin of the proof pages pinpointing the source of the question and/or comment. The numbers will be removed from the margins prior to publication.

- Except for the term "and/or," the use of the slash is discouraged between words and abbreviations, as the intent of the solidus is ambiguous. Several possibilities for its meaning exist, among them "and," "or," "and/or," and "plus." We ask that more precise, and therefore more meaningful, conjunctions be used. For terms that are diagrammatically opposed, we use a hyphen (e.g., liquid-solid interface, vacancy-acceptor interface).
- 2 It is journal style to break down fractions in superscripts and subscripts. Please check that fractions and added bracketing in Eqs. (6),(7),(9) appear acceptably.
- 3 Please write out FWM. Was FWHM intended?
- Please review the funding information section of the proof's cover letter and respond as appropriate. We must receive confirmation that the funding agencies have been properly identified before the article can publish.
- S NOTE: External links, which appear as blue text in the reference section, are created for any reference where a Digital Object Identifier (DOI) can be found. Please confirm that the links created in this PDF proof, which can be checked by clicking on the blue text, direct the reader to the correct references online. If there is an error, correct the information in the reference or supply the correct DOI for the reference. If no correction can be made or the correct DOI cannot be supplied, the link will be removed.
- A check of online databases revealed a possible error in Ref. [30]. The year has been changed from '2016' to '2017'. Please confirm this is correct.

# **Titles in References**

The editors now encourage insertion of article titles in references to journal articles and e-prints. This format is optional, but if chosen, authors should provide titles for *all* eligible references. If article titles remain missing from eligible references, the production team will remove the existing titles at final proof stage.

# ORCIDs

Please follow any ORCID links ((b)) after the authors' names and verify that they point to the appropriate record for each author.

## **Funding Information**

Information about an article's funding sources is now submitted to Crossref to help you comply with current or future funding agency mandates. Crossref's Open Funder Registry (https://www.crossref.org/services/funder-registry/) is the definitive registry of funding agencies. Please ensure that your acknowledgments include all sources of funding for your article following any requirements of your funding sources. Where possible, please include grant and award ids. Please carefully check the following funder information we have already extracted from your article and ensure its accuracy and completeness:

• Ministry of Education - Singapore, FundRef ID http://dx.doi.org/10.13039/501100001459 (Republic of Singapore/SG)

### **Other Items to Check**

- Please note that the original manuscript has been converted to XML prior to the creation of the PDF proof, as described above. Please carefully check all key elements of the paper, particularly the equations and tabular data.
- Title: Please check; be mindful that the title may have been changed during the peer-review process.
- Author list: Please make sure all authors are presented, in the appropriate order, and that all names are spelled correctly.
- Please make sure you have inserted a byline footnote containing the email address for the corresponding author, if desired. Please note that this is not inserted automatically by this journal.
- Affiliations: Please check to be sure the institution names are spelled correctly and attributed to the appropriate author(s).
- Receipt date: Please confirm accuracy.
- Acknowledgments: Please be sure to appropriately acknowledge all funding sources.
- References: Please check to ensure that titles are given as appropriate.
- Hyphenation: Please note hyphens may have been inserted in word pairs that function as adjectives when they occur before a noun, as in "x-ray diffraction," "4-mm-long gas cell," and "*R*-matrix theory." However, hyphens are deleted from word pairs when they are not used as adjectives before nouns, as in "emission by x rays," "was 4 mm in length," and "the *R* matrix is tested."

Note also that Physical Review follows U.S. English guidelines in that hyphens are not used after prefixes or before suffixes: superresolution, quasiequilibrium, nanoprecipitates, resonancelike, clockwise.

- Please check that your figures are accurate and sized properly. Make sure all labeling is sufficiently legible. Figure quality in this proof is representative of the quality to be used in the online journal. To achieve manageable file size for online delivery, some compression and downsampling of figures may have occurred. Fine details may have become somewhat fuzzy, especially in color figures. The print journal uses files of higher resolution and therefore details may be sharper in print. Figures to be published in color online will appear in color on these proofs if viewed on a color monitor or printed on a color printer.
- Overall, please proofread the entire *formatted* article very carefully. The redlined PDF should be used as a guide to see changes that were made during copyediting. However, note that some changes to math and/or layout may not be indicated.

# Ways to Respond

- Web: If you accessed this proof online, follow the instructions on the web page to submit corrections.
- *Email:* Send corrections to aps-robot@luminad.com. Include the accession code LQ17413 in the subject line.
- Fax: Return this proof with corrections to +1.855.808.3897.

# If You Need to Call Us

You may leave a voicemail message at +1.855.808.3897. Please reference the accession code and the first author of your article in your voicemail message. We will respond to you via email.

# PHYSICAL REVIEW LETTERS VOL..XX, 000000 (XXXX)

#### Spectral Compression of Narrowband Single Photons with a Resonant Cavity

Mathias A. Seidler,<sup>1</sup> Xi Jie Yeo,<sup>2</sup> Alessandro Cerè<sup>1</sup>,<sup>1</sup> and Christian Kurtsiefer<sup>1,2,\*</sup>

<sup>1</sup>Centre for Quantum Technologies, National University of Singapore, 3 Science Drive 2, Singapore 117543 <sup>2</sup>Department of Physics, National University of Singapore, 2 Science Drive 3, Singapore 117551

(Received 19 March 2020; accepted 7 October 2020)

We experimentally demonstrate a spectral compression scheme for heralded single photons with narrow spectral bandwidth around 795 nm, generated through four-wave mixing in a cloud of cold <sup>87</sup>Rb atoms. The scheme is based on an asymmetric cavity as a dispersion medium and a simple binary phase modulator, and can be, in principle, without any optical losses. We observe a compression from 20.6 MHz to less than 8 MHz, almost matching the corresponding atomic transition.

12

DOI:

1

2

3

4

5

6 7

8

9 10

11

Introduction.-Efficient atom-light interactions at the 13 single quantum level is at the core of several proposals 14 15 for storing, processing, and relaying quantum information [1-4]. Many of these schemes require single "flying" 16 17 photons to match the spectrum of atomic transitions [5–8]. Single photons can be emitted from trapped ions 18 [9,10], atoms [11–13], or solid-state systems [14–16]. 19 However, the spectral width of the generated photons 20 may not always match the spectral width of the receiving 21 22 systems. Therefore, methods to engineer the photon spectrum may be required. 23

The simplest method for this is to passively filter the 24 spectrum of bright broadband sources [17,18], with a 25 sometimes significant reduction of brightness, making 26 photon-atom interaction experiments that require a high 27 interaction rate [19,20] difficult. More advanced methods to 28 29 manipulate the spectrum of single photon sources to match that of atomic transitions include restricting the spectral 30 31 mode of emitters with cavities [11,16,21,22], or using electromagnetically induced transparency in atomic ensem-32 bles [23,24] in the source mechanism altogether. As 33 spectral filtering or engineering of the photon generation 34 35 mechanism may not always be possible, it would be 36 desirable to modify the spectrum of a given photon source while maintaining the brightness. To our knowledge, the 37 only experiments to modify the photon spectrum of 38 narrowband single photons use gradient echo quantum 39 40 memories [25,26]. However, this was only demonstrated for photons with spectral bandwidths narrower than atomic 41 absorption linewidths. 42

Here, we demonstrate an alternative technique that 43 compresses the spectral bandwidth of single photons with 44 a spectral bandwidth a few times broader than the corre-45 sponding atomic absorption linewidth, while in principle, 46 maintaining the photon rates. The technique is based on the 47 ideas of time lenses invented for temporal imaging [27,28], 48 49 where the temporal and spectral characteristics of ultrafast electromagnetic pulses [29-31] are manipulated. It turns 50

out that single photon states can be manipulated in a similar way, complementing the techniques for lossless temporal envelope manipulation of narrowband single photon states demonstrated in [32,33].

51

52

53

54

55

56

57

58

59

60

61

62

63

64

65

66

67

68

69

70

71

72

73

74

75

76

77

78

79

80

81

82

83

84

85

86

87

88

Spectral compression of single photon wave packets is achieved in two steps. First, the wave packet is spread out in time such that the width of its envelope is compatible with a narrow spectrum; this can be done using a dispersive element that spreads out different frequency components of the wave packet in time, effectively generating a chirped wave packet. In the second step, a time-dependent phase shift is applied. This step changes the spectral energy distribution of the wave packet.

Previous time-lens-based spectral compression schemes were performed on ultrashort pulses, using optical fibers or diffraction gratings as dispersive elements [29-31]. The suitability of a dispersive element for a spectral compression scheme is related to the spectral bandwidth of the optical pulse. The bandwidths of the ultrashort pulses used in previous time-lens-based spectral compression schemes are typically on the order of 0.1, ..., 1 THz, and the length of optical fibers used to generate a significant temporal broadening of these pulses are on the order of 0.1, ..., 10 km. However, photonic wave packets interacting with single emitters like atoms or molecules have spectral bandwidths on the order of MHz, which would require optical fibers on the order of  $10^8$  m to generate a suitable temporal broadening for spectral compression. Transmitting light through such a long fiber would not only be impractical, but also prohibitively lossy. Similarly, currently available gratings would not be able to significantly disperse photonic wave packets with bandwidths of a few MHz. We overcome this problem by using the dispersive properties of an optical cavity instead. While the dispersion in optical cavities can be much larger, the process then requires a different time-dependent phase shift in the second step to complete the spectral compression process.

89 Theory.-To understand the spectral compression scheme, we start with an initial single photon wave packet, 90 described by an envelope  $|\psi(t)|^2$  of its intensity in time, and 91 its corresponding power spectrum  $|\Psi(\omega; \omega_0, \Gamma_p)|^2$ , con-92 nected by the Fourier transform  $F: \Psi(\omega) = F[\psi(t)]$ . The 93 94 nearly monochromatic wave packet shall be characterized by a central frequency  $\omega_0$  and a spectral width  $\Gamma_p$ . The 95 spreading out of the wave packet in time is accomplished 96 97 Í by reflection off an asymmetric cavity, with an input-output coupler with a low transmission, and a second high-98 reflective mirror, similar to the setup used in [34]. 99

100 If the losses in the cavity are negligible compared to the 101 transmission of the coupling mirror, and the cavity line-102 width  $\Gamma_c$  and photon bandwidth  $\Gamma_p$  are much smaller than 103 free spectral range of the cavity, the action of the cavity to a 104 wave packet near its resonance  $\omega_c$  can be described by a 105 transfer function

$$C(\omega;\omega_c,\Gamma_c) \approx -\frac{\Gamma_c + i2(\omega - \omega_c)}{\Gamma_c - i2(\omega - \omega_c)},$$
(1)

106 which modifies the incoming spectral wave packet 108  $\Psi(\omega; \omega_0, \Gamma_p)$  to a new one,

$$\Psi'(\omega;\Delta\omega,\Gamma_c,\Gamma_p) = \Psi(\omega;\omega_0,\Gamma_p)C(\omega;\omega_c,\Gamma_c), \quad (2)$$

109 where  $\Delta \omega = \omega_0 - \omega_c$  is the detuning between the wave 111 packet and the cavity resonance. For a lossless cavity, this 112 wave packet has the same power spectrum as  $\Psi(\omega)$  because 113  $|C(\omega; \omega_c, \Gamma_c)|^2 = 1$ . The temporal envelope of the reflected 114 wave packet, obtained through the inverse Fourier trans-115 form  $F^{-1}$ ,

$$\psi'(t;\Delta\omega,\Gamma_c,\Gamma_p) = F^{-1}[\Psi'(\omega;\Delta\omega,\Gamma_c,\Gamma_p)], \quad (3)$$

116 is now broader in time, and has acquired a time-dependent 118 phase  $\phi'(t; \Delta \omega, \Gamma_p, \Gamma_c)$ .

Similar to Fourier-transform limited pulses, where the time-bandwidth product is minimized by a frequencyindependent spectral phase, we can reduce the spectral bandwidth of the heralded single photon by removing any time-dependent phase. This is done by applying a timedependent phase shift

$$\phi_e(t) = -\phi'(t; \Delta\omega, \Gamma_p, \Gamma_c), \qquad (4)$$

resulting in the spectrally compressed wave packet

$$\psi''(t;\Delta\omega,\Gamma_p,\Gamma_c) = \psi'(t;\Delta\omega,\Gamma_p,\Gamma_c)e^{i\phi_e(t)}.$$
 (5)

129 To quantify the compression, we compare the spectral 130 widths before and after the compression obtained from the 131 respective power spectrum  $|\psi''(\omega; \Delta \omega, \Gamma_p, \Gamma_c)|^2$  obtained 132 through a Fourier transform of Eq. (5). We now consider the specific case of a heralded single 133 photon emerging from an atomic cascade decay, where we 134 intend to compress the idler photon (see inset of Fig. 2). 135 Detection of a signal photon projects the field in the idler 136 mode into the heralded state 2 137

$$\psi(t) = \sqrt{\Gamma_p} e^{-\Gamma_p / [2(t-t_0)]} \Theta(t-t_0), \qquad (6)$$

where  $t_0$  and t are the detection times of the signal and idler photons, respectively. The exponential decay with the constant  $\Gamma_p$  is a characteristic of the spontaneous process, while the Heaviside step function  $\Theta$  is a consequence of the well-defined time order of the cascade decay process. For simplicity, we set  $t_0 = 0$ .

This temporal profile corresponds to a Lorentzian power 145 spectrum for the idler photons, and its bandwidth is 146 described by the full width at half maximum  $\Gamma_p$ , which 147 also corresponds to the spectral window containing 50% of 148 the total pulse energy. However, the compressed spectrum 149  $|F^{-1}[\psi''(t)]|^2$  has multiple maxima, and is distinctly differ-150 ent from distributions where the full width at half maximum 151 naturally quantifies the bandwidth. Hence, we instead define 152 bandwidth as the smallest spectral width containing 50% of 153 the total pulse energy, as this definition of bandwidth is 154 compatible for both a Lorentzian and a generic spectrum. 155

To obtain the optimal cavity parameters, we numerically 156 minimize the bandwidth of the compressed photon spectrum. We find that the maximal compression is achieved by 158 a resonant cavity  $\Delta \omega = 0$  with a bandwidth of  $\Gamma_c \approx \Gamma_p/4$ . 159 Under these conditions, the compressed single photon time 160 envelope can be written as 161

$$\psi''(t) = e^{-i\phi'(t)} \sqrt{\Gamma_p} \frac{2\Gamma_c e^{-(\Gamma_c/2)t} - (\Gamma_p + \Gamma_c)e^{-(\Gamma_p/2)t}}{\Gamma_p - \Gamma_c} \Theta(t),$$
(7)

with a phase function

$$\phi'(t) = \pi \Theta \left( t - 2 \frac{\log(\frac{\Gamma_p + \Gamma_c}{2\Gamma_c})}{\Gamma_p - \Gamma_c} \right).$$
(8)

This is a step function changing the phase by  $\pi$ , with the transition occurring at the minimum of the dispersed photon's temporal intensity profile. The narrowest bandwidth achievable with compression based on an asymmetric cavity with this strategy is  $\sim 0.3\Gamma_p$ ; the temporal envelopes and power spectra shown in Fig. 1 correspond to this choice. 171

*Experiment.*—Details of the actual experiment are shown in Fig. 2. We generate the time-ordered photon pairs by four-wave mixing in a cold ensemble of <sup>87</sup>Rb atoms in a cascade level scheme [12]. Pump beams at 780 and 776 nm excite atoms from the  $5S_{1/2}$ , F = 2 ground level to the  $5D_{3/2}$ , F = 3 level via a two-photon transition. The 762 nm (signal) and 795 nm (idler) photon pairs emerge from a 172

163



F1:1 FIG. 1. Concept of spectral compression. The top row shows F1:2 temporal intensity profiles  $|\psi(t)|^2$  in various stages of the spectral F1:3 compression, the bottom row the corresponding power spectra F1:4  $|\Psi(\omega)|^2$ . The initial pulse is dispersed by a cavity, leading to a F1:5 new temporal shape, but an unchanged spectrum. An electrooptical modulator (EOM) manipulates the phase  $\phi'(t)$  of the pulse F1:7 which leads to a narrower spectrum.

cascade decay back to the ground level, and are coupled to 179 180 single mode fibers. Phase matching is ensured with all four modes propagating collinearly in the same direction. The 181 two pumps have a focus in the cloud with a beam waist of 182 183 about 400  $\mu$ m. The 780 nm pump is 55 MHz blue detuned from the  $5S_{1/2}$ , F = 2 to  $5P_{3/2}$ , F = 3 transition and has an 184 optical power of 0.25 mW. The 776 nm pump has an optical 185 power of 11.4 mW, and is tuned such that the two-photon 186 transition to the  $5D_{3/2}$ , F = 3 state is 5 MHz blue detuned. 187 188 When the excited atoms decay via the  $5D_{1/2}$ , F = 2 state back into the initial ground state, photons with a wave-189 length of 762 nm and 795 nm photon are emitted [12]. 190



F2:1FIG. 2. Schematic setup for generation and spectral compres-<br/>sion of heralded single photons.  $D_{S,I}$ : single-photon detectors;F2:3EOM: electro-optical modulator; PBS: polarizing beam splitter;F2:4QWP: quarter-wave plate; IF: interference filter. Inset: energyF2:5level scheme for four-wave mixing in  ${}^{87}$ Rb.

After suppressing residual pump light and separating 191 signal and idler photons into different modes, we collect 192 them into single mode fibers. The 762 nm signal photons 193 are detected with an avalanche photo diode and herald the 194 presence of 795 nm idler photons. The time correlation 195 between the detection in the signal and idler modes (open 196 circles in Fig. 3) correspond to the envelope  $|\psi(t)|^2$  of the 197 intensity in time. 198

199

200

201

202

203

204

205

206

207

208

209

210

211

212

213

214

215

216

217

We measure the initial power spectrum of the wave packet (open circles in Fig. 4) by correlating it with a the photon rate transmitted through a Fabry-Perot cavity (FP) with linewidth  $\Gamma_{\rm FP} \approx 2\pi \times 2.6$  MHz. The transmission is recorded at different detunings of the cavity from the atomic resonance. The observed spectrum was observed to have a full width at half maximum of 20(2) MHz, wider than the atomic linewidth of 6 MHz due to collective emission effects in the cloud [35,36].

The 795 nm idler photons are then coupled to the dispersion cavity, with a coupling mirror of nominal reflectivity  $R_1 = 0.97$ , and a high reflector with  $R_2 = 0.9995$  separated by 10.1 cm, corresponding to a free spectral range of 1.48 GHz, and a measured linewidth  $\Gamma_c \approx 2\pi \times 7.3$  MHz. A Pound-Drever-Hall frequency lock keeps the cavity resonant to the central frequency of the photons throughout the experiment. The measured time envelope of the single photon wave packet after dispersion is shown as filled dots in Fig. 3.

The spectral compression is completed by applying the218temporal phase of Eq. (8), in the form of a phase switch219synchronized to the photon passage through a fiber con-220nected electro-optical modulator (EOM). Since the idler221photon is heralded, we use the detection of the signal222photon as a reference signal for triggering the phase switch223



FIG. 3. Detection time distribution for the heralded photon before (open blue circles) and after (filled red dots) the dispersion cavity. We fit an exponential decay Eq. (6) to the initial time correlation (blue solid line), from which we infer the photon bandwidth  $\Gamma_p$ . The simulated temporal profile after the photon passed through the dispersion cavity [red line, calculated from Eq. (2)], matches our experimental data (filled dots) well. F3:1 F3:2 F3:2 F3:3 F3:4 F3:5 F3:6 F3:6 F3:7



F4:1 FIG. 4. Spectral profile of heralded photons before (blue) and F4:2 after (red) spectral compression, obtained by measuring the F4:3 photon transmission rate through the Fabry-Perot cavity at F4:4 different cavity detunings. The solid lines are calculated from F4:5 Eq. (5), with  $\Gamma_p$  inferred from the temporal envelope measure-F4:6 ment of the photons from the source, and  $\Gamma_c$  by experimentally F4:7 characterizing the cavity bandwidth. Shaded areas cover 50% of F4:8 the total power for each spectrum.

224 after an appropriate time delay. Our dispersed photon is 225 approximately 80 ns long, which corresponds to a spatial 226 spread of 16 m in a fiber with a refractive index of 1.5. The 227 phase modulator has an active length of 90 mm, so at any instant only a small part of the photon resides inside the 228 EOM, and we are able to modulate the two parts of 229 230 the photon with different phases. The correct timing 231 of the phase modulation is ensured by measuring the length of the fibers and the electric signal lines with a 232 timing uncertainty < 0.5 ns, on par with the electrical rise 233 time of the phase change signal. This order of timing 234 235 uncertainty can be tolerated, since the majority of second 236 photon wave packet part (Fig. 3, blue line from 15 to 100 ns) gets a phase shift of  $\pi$ . The phase flip is applied 237 238 right after the first part of the dispersed photon exits the modulator, and the second part starts to propagate through 239 240 it. This timing is indicated as the dashed line in Fig. 1. We finally measure the compressed photon spectrum by again 241 recording the photon transmission rate through the Fabry-242 Perot cavity, shown as filled dots in Fig. 4. 243

To obtain an initial photon bandwidth  $\Gamma_p$ , we fit the 244 245 decaying exponential term in Eq. (6) to the observed coincidence probability (open circles in Fig. 3). The solid 246 red line in Fig. 3 corresponds to an expected temporal 247 profile of the photon after the dispersion cavity, calculated 248 from Eq. (7), with an inferred photon bandwidth 249  $\Gamma_p = 2\pi \times 20.6(2)$  MHz obtained from the fit of the initial 250 photon shape, and the cavity linewidth  $\Gamma_c \approx 2\pi \times 7.3$  MHz 251 measured earlier. The observed temporal envelope after the 252 dispersion cavity (full dots in Fig. 3) agrees very well with 253 254 the expected profile.

The measured spectral profiles before and after compression are shown in Fig. 4. The spectrum of the uncompressed photons (open circles) exhibits a dip around 257 the central frequency, which was also observed without the 258 compression optics. We attribute this to reabsorption of the 259 generated photons by the atomic cloud. We model this 260 spectrum  $S(\omega)$  by considering two processes: First, we 261 consider the spectrum  $P(\omega)$  of the photon emitted by the 262 atomic cloud which can be obtained by the product of the 263 spectrum of the photon produced by our FWM process 3 264  $L(\omega;\Gamma_p) = (2/\pi)(\Gamma_p/4\omega^2 + \Gamma_p^2)$  and an absorption term 265 describing the attenuation of the photon by our atomic 266 cloud of optical density OD 267

$$P(\omega; \text{OD}, \Gamma_n, \Gamma_a) = AL(\omega; \Gamma_n) e^{-\text{OD}[\Gamma_a^2/(4\omega^2 + \Gamma_a^2)]}.$$
 (9)

The scaling factor A is used to account for the detected 269 coincidence rate, and  $\Gamma_a$  is the spectral width of 270 the absorption feature. From our fit, we extract 271  $\Gamma_a = 2\pi \times 1.38(1.6)$  MHz, which does not correspond to 272 the absorption linewidth of the corresponding atomic 273 linewidth ( $2\pi \times 5.7$  MHz) of the  $5S_{1/2} \rightarrow 5P_{1/2}$  transition. 274 Further work is necessary to understand this observation. 275 The inferred photon bandwidth  $\Gamma_p = 2\pi \times 20.6(2)$  MHz 276 was determined from the photon coincidence time corre-277 lation (Fig. 3). Second, we consider the effect of the 278 Fabry-Perot cavity used to sample the spectral profile by 279 convolution the above result with  $L'(\omega; \Gamma_{\rm FP}) =$ 280  $[\Gamma_{\rm FP}^2/(4\omega^2+\Gamma_{\rm FP}^2)]$  to model the observed spectrum: 281

$$S(\omega) = (P * L')(\omega). \tag{10}$$

We fit the model  $S(\omega)$  (Fig. 4, black line) to the exper-283 imental data corresponding to the spectral profile of the 284 photon without sending a signal to the EOM used to 285 compress the photon. The model, without considering the 286 attenuation of the atomic cloud, is given by  $(AL * L')(\omega)$ 287 (Fig. 4, blue line). This is provided as a reference for a 288 Fourier-transform limited photon with an exponentially 289 decaying envelope, as emitted by a single atom-the 290 scenario examined in the theory section. 291

To apply the analysis in the theory section for predicting 292 the spectrum of the compressed photon, we first rescale the 293 power spectrum  $|\Psi''(\omega)|^2$ , calculated from Eq. (7), with A, 294 which was extracted from the previous fit. Then, we 295 convolve this spectrum with  $L'(\omega; \Gamma_{\rm FP})$  to obtain the 296 expected compressed photon spectrum  $(A|\Psi''|^2 * L')(\omega)$ . 297 Figure 4 (red line) shows the modeled power spectrum 298 slightly deviating from the measured result (red dots), 299 exhibiting a lower peak coincidence rate where an absorp-300 tion dip occurs in the uncompressed photon spectrum (blue 301 dots). We attribute this difference to the fact that our model 302 does not fully account for the effects imposed by the 303 atomic cloud. 304

*Discussion.*—By definition, spectral compression 305 reduces the width of a spectral distribution, resulting in 306 an increased photon rate and intensity at the central 307 308 frequency. In our experiment, we observed a bandwidth of 20(2) MHz for the initial photon, and 8(2) MHz for the 309 compressed photon from spectroscopy using a 2.6 MHz 310 311 cavity. This almost matches the natural D transition linewidth of 6 MHz in <sup>87</sup>Rb. The maximal photon transmission 312 through the spectroscopic cavity is increased by a factor of 313 2.39(4), indicating a successful spectral compression of 314 narrowband photons. 315

The compressions mechanism is, in principle, lossless since both cavity and phase modulators can have arbitrarily low losses. In our experiment, however, we observe an overall transmission of 22% through the compression optics; the dispersion optics alone (PBS, QWP, dispersion cavity) has a transmission of 72%, and the fiber-based EOM a transmission of 30% including the fiber coupling.

To compare the compression method with simple passive 323 filtering, we calculate the transmission T of the 20 MHz 324 325 bandwidth photons produced by our source through both bandwidth-limiting schemes, and an analyzer cavity with a 326 bandwidth of 6 MHz bandwidth and resonant with the 327 central frequency of the input photons to model an atomic 328 absorption process corresponding to the  $5S_{1/2} \rightarrow 5P_{1/2}$ 329 transition in <sup>87</sup>Rb. With a lossless compression system, we 330 find T = 44%, while a resonant filter cavity of the same 331 bandwidth of 6 MHz leads to T = 14%, illustrating the 332 advantage of the compression method. By replacing the 333 fiber-based EOM with a free-space EOM with an optical 334 transmission > 95%, the compression method would 335 significantly surpass the transmission of a passive filter. 336

Optimal spectral compression of a photon with band-337 width  $\Gamma_p$  in the cavity-based scheme is achieved if the 338 dispersion cavity has a bandwidth of  $0.25\Gamma_p$ . Since the 339 340 amount of spectral compression is limited by the dispersion mechanism, dispersion engineering of structured dielectric 341 342 media [37–39] or multiple combined optical cavities may allow us to further increase the spectral compression. This 343 method is not limited to the atomic system in our experi-344 345 ment-it can be adapted to a wide range of wavelengths and spectral widths, and therefore even allow us to match 346 the spectral properties to different types of quantum 347 systems, e.g., in a hybrid quantum network [40]. 348

We thank Adrian Nugraha Utama for useful discussions
on the theoretical modeling and Jianwei Lee for his
valuable input while writing this script. This work was
supported by the Ministry of Education in Singapore.

<sup>\*</sup>christian.kurtsiefer@gmail.com

355 354 356

- 357 [1] J. I. Cirac, P. Zoller, H. J. Kimble, and H. Mabuchi,
   358 S Phys. Rev. Lett. 78, 3221 (1997).
- 359 [2] H.-J. Briegel, W. Dür, J. I. Cirac, and P. Zoller, Phys. Rev.
  360 Lett. 81, 5932 (1998).
- 361 [3] E. Waks and C. Monroe, Phys. Rev. A 80, 062330 (2009).
- 362 [4] H. J. Kimble, Nature (London) **453**, 1023 (2008).

[5] T. Wilk, S. C. Webster, A. Kuhn, and G. Rempe, Science 317, 488 (2007). 363

364

365

366

367

368

369

370

371

372

373

374

375

376

377

378

379

380

381

382

383

384

385

386

387

388

389

390

391

392

393

394

395

396

397

398

402

403

404

405

406

407

408

409

410

411

412

413

414

415

416

419

420

421

422

- [6] K. Hammerer, A. S. Sørensen, and E. S. Polzik, Rev. Mod. Phys. 82, 1041 (2010).
- [7] M. D. Lukin, Rev. Mod. Phys. 75, 457 (2003).
- [8] M. Steiner, V. Leong, M. A. Seidler, A. Cerè, and C. Kurtsiefer, Opt. Express 25, 6294 (2017).
- [9] M. Keller, B. Lange, K. Hayasaka, W. Lange, and H. Walther, Nature (London) **431**, 1075 (2004).
- [10] M. Almendros, J. Huwer, N. Piro, F. Rohde, C. Schuck, M. Hennrich, F. Dubin, and J. Eschner, Phys. Rev. Lett. 103, 213601 (2009).
- [11] A. Kuhn, M. Hennrich, and G. Rempe, Phys. Rev. Lett. 89, 067901 (2002).
- [12] B. Srivathsan, G. K. Gulati, B. Chng, G. Maslennikov, D. N. Matsukevich, and C. Kurtsiefer, Phys. Rev. Lett. 111, 123602 (2013).
- [13] J. Park, H. Kim, and H. S. Moon, Phys. Rev. Lett. 122, 143601 (2019).
- [14] C. Kurtsiefer, S. Mayer, P. Zarda, and H. Weinfurter, Phys. Rev. Lett. 85, 290 (2000).
- [15] P. Michler, A. Kiraz, C. Becher, W. Schoenfeld, P. Petroff, L. Zhang, E. Hu, and A. Imamoglu, Science 290, 2282 (2000).
- [16] E. Moreau, I. Robert, J. Gérard, I. Abram, L. Manin, and V. Thierry-Mieg, Appl Phys. Lett. 79, 2865 (2001).
- [17] E. Meyer-Scott, N. Montaut, J. Tiedau, L. Sansoni, H. Herrmann, T. J. Bartley, and C. Silberhorn, Phys. Rev. A 95, 061803(R) (2017).
- [18] C. Schuck, F. Rohde, N. Piro, M. Almendros, J. Huwer, M. W. Mitchell, M. Hennrich, A. Haase, F. Dubin, and J. Eschner, Phys. Rev. A 81, 011802(R) (2010).
- [19] L.-M. Duan, M.D. Lukin, J.I. Cirac, and P. Zoller, Nature (London) 414, 413 (2001).
- [20] B. B. Blinov, D. L. Moehring, L.-M. Duan, and C. Monroe, Nature (London) 428, 153 (2004).
- [21] J. McKeever, A. Boca, A. D. Boozer, R. Miller, J. R. Buck,
   A. Kuzmich, and H. J. Kimble, Science 303, 1992 (2004).
   [22] F. Wolfgramm, X. Xing, A. Cerè, A. Predojević, A.M.
   401
- [22] F. Wolfgramm, X. Xing, A. Cerè, A. Predojević, A. M. Steinberg, and M. W. Mitchell, Opt. Express 16, 18145 (2008).
- [23] M. Eisaman, A. André, F. Massou, M. Fleischhauer, A. Zibrov, and M. Lukin, Nature (London) 438, 837 (2005).
- [24] L. Zhu, X. Guo, C. Shu, H. Jeong, and S. Du, Appl Phys. Lett. 110, 161101 (2017).
- [25] B. Buchler, M. Hosseini, G. Hétet, B. Sparkes, and P. K. Lam, Opt. Lett. 35, 1091 (2010).
- [26] B. M. Sparkes, M. Hosseini, C. Cairns, D. Higginbottom, G. T. Campbell, P. K. Lam, and B. C. Buchler, Phys. Rev. X 2, 021011 (2012).
- [27] B. H. Kolner and M. Nazarathy, Opt. Lett. 14, 630 (1989).
- [28] B. H. Kolner, IEEE J. Quantum Electron. **30**, 1951 (1994).
- [29] J. Lavoie, J. M. Donohue, L. G. Wright, A. Fedrizzi, and K. J. Resch, Nat. Photonics 7, 363 (2013).
- [30] M. Karpiński, M. Jachura, L. J. Wright, and B. J. Smith, 417
   Nat. Photonics 11, 53 (2017).
- [31] Y. Li, T. Xiang, Y. Nie, M. Sang, and X. Chen, Sci. Rep. 7, 43494 (2017).
- [32] B. Srivathsan, G. K. Gulati, A. Cerè, B. Chng, and C. Kurtsiefer, Phys. Rev. Lett. 113, 163601 (2014).

- [33] O. Morin, M. Körber, S. Langenfeld, and G. Rempe, Phys. 423 424 Rev. Lett. 123, 133602 (2019).
- 425 [34] B. Srivathsan, G. K. Gulati, A. Cerè, B. Chng, and C. Kurtsiefer, Phys. Rev. Lett. 113, 163601 (2014). 426
- [35] H. H. Jen, Phys. Rev. A 85, 013835 (2012). 427
- [36] A. Cerè, B. Srivathsan, G. K. Gulati, B. Chng, and C. 428 Kurtsiefer, Phys. Rev. A 98, 023835 (2018). 429
- 430 [37] E. Istrate and E. H. Sargent, Rev. Mod. Phys. 78, 455 431 (2006).
- [38] X. Li, M. Pu, X. Ma, Y. Guo, P. Gao, and X. Luo, J. Phys. D 51, 054002 (2018).
- [39] J. D. Joannopoulos, S. G. Johnson, J. N. Winn, and R. D. Meade, Photonic Crystals: Molding the Flow of Light, 2nd ed. (Princeton University Press, Princeton, NJ, USA, 2008).
- [40] G. Kurizki, P. Bertet, Y. Kubo, K. Mølmer, D. Petrosyan, P. 438 Rabl, and J. Schmiedmayer, Proc. Natl. Acad. Sci. U.S.A. 439 112, 3866 (2015).

440

432

433

434

435

436

437

441