Photons make very good flying quantum bits (qubits) because of their weak interaction with the environment. The ability to store a qubit is essential for long-distance quantum communication [1, 3]. This works best if the optical properties of the medium match the bandwidth of the qubits. Scientists have achieved a high efficiency quantum memory as well as a single-photon-level memory in a Λ-type atomic transition [4–6]. The quantum information can be encoded in a polariton [7], which is in a more general sense a coherent superposition of light and matter. The transfer to a purely matter-like excitation is coherent and reversible, the quantum information can be mapped back onto a light field. To implement in addition quantum information processing, strong interactions between stored qubits are required. The atomic excitation involving a high-lying Rydberg state matches these requirements, as it allows for storage, control and retrieval of optical photons via Rydberg polaritons [8].

Rydberg-EIT (electromagnetically induced transparency) in thermal vapors has first been studied in Ref. 17 without any notion of an Rydberg-Rydberg interaction. For the direct observation of interacting Rydberg atoms at room temperature, a bandwidth-limited pulsed excitation was applied to determine interaction strengths corresponding to a few GHz, much stronger than the decoherence rate set by the motion-induced Doppler effect [18, 19]. Many-body phenomena based on this interaction like aggregation [20, 21] or optical bistabilities [22] have been studied in thermal vapor cells. If one adds a de-excitation laser before the Rydberg states do some unwanted stuff like aggregation etc., the corresponding FWM scheme provides the basis for a deterministic single-photon source [23]. It is important that the polariton lifetime, or better said the coherence time, is sufficient to produce a mixed wave fully coherent. The Rydberg polariton lifetime can be limited by dephasing, e.g. due to inhomogeneous differential light shifts in an optical dipole trap for cold atoms [24]. In thermal vapors, dipole traps are not applicable but the lifetime is now limited by motional dephasing. The corresponding bandwidth determines the blockade radius for a given van der Waals interaction.

In this Letter, we investigate the properties of thermal Rydberg polaritons in the absence of Rydberg-Rydberg interactions by a double-pulsed FWM transition. Some of the incident photons are first stored as matter-like polaritons. A pulsed retrieval laser converts the excitations back into light-like polaritons, i.e. photons which we detected. The retrieval laser is temporally delayed in order to probe the effective lifetime of the polaritons. We present measurements for different storage times of Rydberg polaritons, and obtain the lifetime of polaritons to be 1.2 ns for our given bandwidth of the excitation sequence. We detect the retrieved photons produced via FWM by single-photon counters as we only address quite small atomic ensemble. The retrieval light field shows motion-induced revivals, which can be attributed to the constructive interference between the coherent evolution of different velocity classes [25, 26]. All these features can be reproduced by numerical simulations considering the contribution of a whole Doppler ensemble. It is important to note that the temporal evolution can only be explained by a collective response of the ensemble and not by a single-atom master equation. Such a collective behavior is also necessary for the realization of a single photon source as detailed in [24]. Rydberg interactions
generating the FWM signals at \( \sim p \) pulsed retrieval field with wavelength of 475 nm laser of FWHM \( \sim 2.5 \) ns, driving a resonant transition to the Rydberg state \( \vert 4 \rangle \), from where the four-wave mixing process is closed by emitting photons at \( \sim 780 \) nm. The 795 nm laser is focused onto the cell with a 1/e² beam diameter of around 35(5) \( \mu \)m. Both blue beams are overlapped by a pinhole which is imaged into the cell to provide a homogeneous beam profile among the interaction regime at a diameter of \( \sim 67(5) \) \( \mu \)m. We determine the peak Rabi frequencies applied in this experiment to be \( \Omega_{795}/2\pi = 85(10) \) MHz, \( \Omega_{475}/2\pi = 160(10) \) MHz, and \( \Omega_{480}/2\pi = 130(10) \) MHz. More details can be found in Ref. [28].

The time-evolution of the atom-light system can be calculated by a basic four-level transition model coupled by three light fields [25]. We numerically solve the Lindblad equation with the Hamilton operator in a four-level diamond configuration, \( \frac{\partial \rho_v}{\partial t} = -\frac{i}{\hbar} \left( \hat{H}_v, \rho_v \right) + L(\rho_v) \) for the velocity group \( v \). The Lindblad operator \( L(\rho_v) \) includes the various decays [29]. The corresponding Hamiltonian with rotating-wave approximation is

\[
\hat{H}_v = \hbar \begin{pmatrix}
0 & \Omega_{795}(r) & 0 & 0 \\
\Omega_{795}(r) & 0 & \Omega_{480}(r,t) & 0 \\
0 & \Omega_{480}(r,t) & 0 & \Omega_{475}(r,t) \\
0 & 0 & \Omega_{475}(r,t) & 0 \\
\end{pmatrix}
\]

(1)

\( \delta_{v,795}, \delta_{v,480} \), and \( \delta_{v,475} \) are the respective Doppler detunings. The spontaneous decay rates from intermedi-
The generated electric field can be filtered out by the serial optical detection system. Taking into account the apertures at the imaging system including two lenses and one pinhole, we derive the detected electric field after pinhole $E_{\text{out}}(r, t)$ by applying the Fresnel approximation and phase transformation function right behind lenses $E(r, t) e^{-i k_{\text{780}} r^2/2 f}$ ( $f$ is the focal length and $k_{\text{780}}$ is the wave-vector of the signal light) \cite{51}. By this method, we get the time-dependent power $P(t)$ and photon number

$$N_{\text{ph}} = \int P(t) dt / \hbar \omega. \quad (4)$$

$r_{\text{hole}} = 5 \mu m$ is the radius of the pinhole.

The experiment begins with a two-photon excitation. Few photons are first stored in the medium as Rydberg polaritons after the pulse is over. The coherence $\rho_{41}$ is then imprinted onto the D$_2$ transition by applying a retrieval pulsed laser at 480 nm after a variable storage time. We first measure the bandwidth of the coherence $\rho_{31}$ by systematically scanning the detuning of the retrieval laser. The storage time is fixed at 3.1 ns. The measured full width at half maximum (FWHM) bandwidth is ~ 500 MHz, which is the convolution of the $\rho_{31}$ lineshape and the spectrum of the retrieval pulse. In the following measurements, the 480 nm laser is locked at resonance frequency.

We then vary the storage times of the Rydberg polaritons. Looking at the dynamics of the retrieved sig-
signals measured at short delay times (Fig. 2), we observe motion-induced signal revivals, as discussed in Refs. 22, 26. The atomic coherences differ in both amplitude and phase for the different velocity classes. After the excitation pulse, the atomic coherences are evolving freely according to their Doppler detunings with phase $\propto \exp(-ik_{780}vt)$. The temporal shape of the oscillatory FWM signals can be attributed to the coherence interference of different atomic velocity classes. The cyan dashed lines are the numerical results from solving the master equation and extracting the coherence $\rho_{41}$. For longer storage times, we observe slowly decaying tails of the retrieved signals. We attribute this to the effect of spontaneous emission which in the simulation is derived from $\Gamma_4 \cdot \rho_{44} \times$ collection efficiency $[30]$. The numerical results including both types of emission, shown as the red solid lines, fit the measurements well.

With 1.0 ns storage time, 9 retrieved photons are detected. For shorter storage times, the signals are dominated by the coherent collective emission; and for longer delay times, the signals are mainly dominated by the spontaneous emission. The cyan and green dashed lines shown in Fig. 2 represent the simulation of the coherent and spontaneous emissions, respectively, with the parameters used in Fig. 2 and the red solid line is the sum of both types of emission. The adjusted density in the simulations is $0.92 \times 10^{15}$ cm$^{-3}$, which is in the range of the experimentally determined density. An exponential fit to the coherent part of the emission (i.e. for storage times less than 6.5 ns) shows a polariton lifetime of 1.2 ns. The lifetime is not dominated by the Rydberg population decay of 6.9 $\mu$s neither by the transient time (the time period that atoms move in and out of the interaction regime is around 64 ns). The lifetime is inferred from the Doppler dephasing. Due to the co-propagating configuration of the excitation laser beams, the effective wave vector to the Rydberg state is $k_{\text{eff}} = k_{795} + k_{475}$. The FWHM bandwidth of Rydberg population is $\Delta v_{\text{coh}} = k_{\text{eff}} \cdot \Delta v$. From the numerical calculation, we know that the velocity band of Rydberg-excited atoms has a FWHM of $\Delta v = 52$ m/s, implying the bandwidth of $\Delta v_{\text{coh}} = 175$ MHz. The corresponding Doppler dephasing lifetime is 0.91 ns, close to the measurements.

In future works, we will increase the Rydberg quantum number to 37 and above, for example, for which interaction energy of up to $\sim 2$ GHz $[19]$ is much larger than the measured excitation bandwidth. Moreover, for these parameters a Rydberg blockade volume of $\sim 6 \mu m^3$ is expected which can be matched by reducing the excitation volume by combining a tightly focused beam and a micrometer-sized cell. Therefore, this scheme can be used for generating non-classical light fields $[22]$.

In summary, we store and retrieve thermal Rydberg polaritons in a bandwidth-limited double-pulsed scheme. With 1.0 ns delay time, we obtain 9 retrieved photons. The thermal Rydberg polaritons have a lifetime of 1.2 ns, almost entirely limited by motional dephasing of the atoms. Our numerical calculation including the spatial profiles of the laser beams and also the light propagation of the generated field fits well to the temporal structures and lifetime of the retrieved signals. We propose that this FWM-excitation scheme combined with strong Rydberg interactions can be used for generating non-classical light fields or manipulating quantum information carriers by photons.

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* Electronic address: f.ripka@physik.uni-stuttgart.de
† Electronic address: t.pfau@physik.uni-stuttgart.de
‡ Electronic address: yhchen920@gmail.com

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