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Creation of two-photon states via interactions between Rydberg atoms during light storage

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We propose a method to create two-photon states in a controllable way using interaction between the Rydberg atoms during the storage and retrieval of slow light. A distinctive feature of the suggested procedure is that the slow light is stored into a superposition of two atomic coherences under conditions of electromagnetically induced transparency. Interaction between the atoms during the storage period creates entangled pairs of atoms in a superposition state that is orthogonal to the initially stored state. Restoring the slow light from this new atomic state one can produce a two-photon state with a second-order correlation function determined by the atom-atom interaction and the storage time. Therefore the measurement of the restored light allows one to probe the atom-atom coupling by optical means with a sensitivity that can be increased by extending the storage time. As a realization of this idea we consider a many-body Ramsey-type technique which involves $\pi/2$ pulses creating a superposition of Rydberg states at the beginning and the end of the storage period. In that case the regenerated light is due to the resonance dipole-dipole interaction between the atoms in the Rydberg states.

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I. INTRODUCTION

Generation of photon pairs has a fundamental and technological significance [1], and can be used in quantum measurement and quantum information transfer [2]. Production of nonclassical photon pairs via the electromagnetically induced transparency (EIT) has been first demonstrated a decade ago [3,4]. Subsequently photon pairs with a controllable profile have been created [5,6] by employing the slow light in a double-Λ system. In related recent developments, nonlinear quantum optics has been investigated for slow light using Rydberg atoms [7–17]. Since the van der Waals interaction between the atoms increases with the principal quantum number as n^{11}, the interaction between the Rydberg atoms is enhanced by many orders of magnitude compared to the interaction between atoms in the ground state [18–21]. The interaction brings neighboring Rydberg atoms out of the resonance destroying the EIT. Consequently the close-by Rydberg atoms absorb the slow light, so photons become antibunched during propagation of light through the atomic medium [7–12,14,16]. Nonclassical photon or atomic states using Rydberg interactions have also been investigated in Refs. [22–24].

Here we propose another way of generation of correlated two-photon states via storage and retrieval of the slow light in the atomic medium. Unlike conventional light storage [25–33], the probe pulse is now stored in a superposition of two atomic states involving the Rydberg levels. During a subsequent evolution the atom-atom interaction produces entangled pairs of atoms in a superposition state that is orthogonal to the initially stored state. Restoring the slow light from this new atomic state one can produce two-photon states with the second-order correlation function determined by the atom-atom interaction and the storage time. Furthermore, measurement of the second-order correlation function of the restored light allows one to probe the interaction potential by optical means, with a sensitivity that can be enhanced by increasing the storage time. Note that the creation of an atomic superposition and a subsequent retrieval of light form the orthogonal superposition represents a Ramsey interferometry [34–38].

Previously regeneration of light from an initially unpopulated coherence was implemented using an external detuning in a tripod atom-light coupling scheme [39] (see also a related work [40]). In that case the regenerated light remains classical by storing a classical light. On the other hand, in the current proposal the detuning is caused by the interaction between close-by Rydberg atoms leading to regeneration of correlated photon pairs.

As a specific realization of the proposed idea we consider in this paper a light-matter interaction in an ensemble of atoms characterized by a ladder scheme of energy levels shown in Fig. 1(a). After switching off the control beam, the probe beam is stored in a coherence between the ground state g and the Rydberg state s. In order to create a superposition of two Rydberg states and to restore the slow light from an orthogonal superposition we propose to apply at the beginning and at the end of the storage the $\pi/2$ optical pulses coupling the Rydberg s and p states.

The paper is organized as follows. In Sec. II we present the proposed setup. In Sec. III we investigate how the stored atomic state is changed by the atom-atom interactions. In Sec. IV we analyze the probe pulse restored from this atomic state. Section V summarizes our findings and discusses possible experimental implementation.

II. FORMULATION

We consider a light-matter interaction in an ensemble of atoms characterized by a ladder scheme of energy levels shown in Fig. 1(a). We assume that the size of the atomic medium is much larger than the optical wavelength. We include the atomic ground s and excited p states labeled, respectively, by $|g\rangle$ and $|e\rangle$, as well as Rydberg s and p states denoted by $|s\rangle$ and $|p\rangle$. The corresponding energies are $\hbar\omega_g$, $\hbar\omega_e$, $\hbar\omega_s$, and $\hbar\omega_p$. The atoms, initially in the ground state $|g\rangle$, are

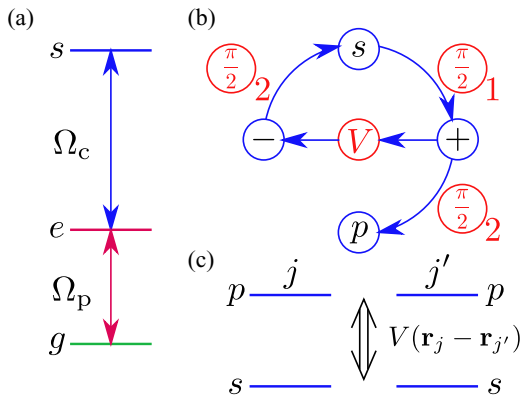

FIG. 1. (a) The atomic ground state $|g\rangle$ is coupled to the excited state $|e\rangle$ by the probe field Ω_p. The latter $|e\rangle$ is coupled to the Rydberg state $|s\rangle$ by the control laser Ω_c. (b) Changes of the atomic states due to the action of the first $\pi/2$ pulse, the RDDI potential V, and the second $\pi/2$ pulse. (c) The RDDI $V = V(|\mathbf{r}_j - \mathbf{r}_{j'}|)$ exchanges of the s and p Rydberg states between nearby atoms j and j'.

illuminated by a probe laser field with a central frequency ω_p. Additionally there is a more intense classical control field with a frequency ω_c. The probe (control) field resonantly drives the atomic transition $g \to e$ ($e \to s$) with a coupling strength characterized by a Rabi frequency Ω_p (Ω_c).

The incident probe field Ω_p represents a long and flat pulse of a classical light, such that, except for a short transient period, it can be considered to be time independent. The second-order correlation function $g_{\rm in}^{(2)}(\tau)$ of such an incident light is constant. We are interested in times τ corresponding to interatomic distances $r = v_{g0}\tau$ much larger than the Rydberg blockade radius, so that the interaction between atoms can be neglected during the propagation of light with a velocity $v_{g0} \ll c$ in the medium. In the previous studies on the nonclassical slow light [7,8,11,12,41] the strong Rydberg blockade during the propagation of slow light provides photons antibunched over a length exceeding the blockade radius. In contrast, the present approach does not rely on the Rydberg blockade, so one can neglect its effects during the propagation of the slow light.

After switching off the control beam, the probe beam is stored in a coherence between the ground state g and the Rydberg state s. Subsequently a $\pi/2$ optical pulse is applied that couples the Rydberg s and p states, as shown in Fig. 1(b). This converts the Rydberg s state into a superposition of the s and p Rydberg states $|+\rangle = (|s\rangle + |p\rangle)/\sqrt{2}$. A similar procedure has been employed in Ref. [13], where a single microwave pulse has been used during the light storage to couple the initial Rydberg state to a neighboring internal state. The medium is then left to evolve freely for a duration T. As we shall see later on, during the storage the resonance dipole-dipole interaction (RDDI) between the atoms in the s and p Rydberg states creates *correlated pairs* of atoms j and j' in an initially unpopulated state $|-\rangle = (|s\rangle - |p\rangle)/\sqrt{2}$ with correlations determined by the RDDI potential $V(|\mathbf{r}_j - \mathbf{r}_{j'}|)$. Note that high fidelity $\pi/2$ microwave pulses should be of a sufficiently large Rabi frequency which exceeds the corresponding strength of the dipole-dipole interaction $\Omega_{\rm rf} > V(r)$ at relevant interatomic distances r.

Just before the retrieval one applies another $\pi/2$ optical pulse coupling the Rydberg s and p states. This converts the state $|+\rangle$ into the Rydberg state $|p\rangle$, whereas the state $|-\rangle$ is transferred back into the Rydberg state $|s\rangle$; see Fig. 1(b). Such a procedure represents a Ramsey-type interferometry involving atom-atom interaction [35–38]. For atoms in the Rydberg state $|p\rangle$ there is no allowed optical transition to the excited state $|e\rangle$. Therefore, when restoring the light, atomic excitations in the s state are converted into probe photons, and the p state excitations remain in the medium. Hence no slow light would be regenerated without the RDDI which converts the internal state $|+\rangle$ into $|-\rangle$ for neighboring atoms. Restoring the probe beam one produces correlated pairs of probe photons, like in the parametric downconversion [42].

Applying the rotating wave approximation [43], a Hamiltonian for the atoms coupled with the laser fields reads in the interaction representation

$$\mathcal{H} = \mathcal{H}_{\rm at-light} + \mathcal{H}_{\rm SP} + \mathcal{H}_{\rm at-at}. \tag{1}$$

Here

$$\mathcal{H}_{\rm at-light} = -\frac{1}{2}\sum_j \left(\Omega_p \sigma_{eg}^j + \Omega_c \sigma_{es}^j + {\rm H.c.}\right) \tag{2}$$

is a Hamiltonian for the atom-light coupling, $\sigma_{ab}^j = |a^j\rangle\langle b^j|$ are (quasi)-spin-flip operators transferring the jth atom from state b to state a [31], with a and b standing for the atomic internal states g, e, s, and p. We have assumed an exact EIT resonance with zero two- and single-photon detunings: $\omega_g + \omega_p + \omega_c - \omega_s = 0$ and $\omega_g + \omega_p - \omega_e = 0$. The term

$$\mathcal{H}_{\rm SP} = \sum_j \Omega_{\rm SP}(t)\left(\sigma_{ps}^j + \sigma_{sp}^j\right) \tag{3}$$

describes the coupling between the Rydberg s and p states due to an external electromagnetic field characterized by a Rabi frequency $\Omega_{\rm SP}(t)$. The latter $\Omega_{\rm SP}(t)$ is composed of two $\pi/2$ pulses, one applied at the beginning of the storage, another one at the end of the storage. At the remaining stages (propagation, storage, and release of light) $\Omega_{\rm SP}(t)$ is off and hence is to be omitted. Finally

$$\mathcal{H}_{\rm at-at} = \sum_{j\neq j'} V(|\mathbf{r}_j - \mathbf{r}_{j'}|)\sigma_{ps}^j \sigma_{sp}^{j'} \tag{4}$$

is the RDDI Hamiltonian leading to exchange of Rydberg states between pairs of atoms, one of which being in the s state and another in the p state. The action of RDDI during the light storage is schematically shown in Fig. 1(c). The strong RDDI between pairs of Rydberg atoms is described by a position-dependent strength $V(|\mathbf{r}_j - \mathbf{r}_{j'}|)$, chosen to be real, where $\mathbf{r}_j$ is a position vector of the jth atom. The condition $j \neq j'$ excludes self-interactions in Eq. (4). Note that a double summation over j and j' ensures that both forward and backward resonance transfer between the states s and p are included in the interaction Hamiltonian (4) which is therefore Hermitian.

III. TIME EVOLUTION OF ATOMIC STATE DURING THE STORAGE OF PROBE PULSE

A. Stored atomic state

When the control field Ω_c is on, the slow light made of dark-state polaritons propagates in the medium with a group velocity $v_{g0} = \Omega_c^2 L/\Gamma\alpha$ [25,28,44,45]. Here α and L are, respectively, an optical density and a length of the medium, whereas Γ is a decay rate of the excited state e. Since the initial probe field is classical, one can replace atomic spin-flip operators σ_{ab}^j by the corresponding density matrix elements $\rho_{ba}^j = \langle\sigma_{ab}^j\rangle$. A duration T of the subsequent light storage is assumed to be much larger than the propagation time of slow light in the medium. Hence the atom-atom interaction has a significant accumulative effect only during the light storage, and one can neglect the interaction effects during the light propagation. Under the EIT condition, the induced coherence between the atomic ground and Rydberg s state $\rho_{sg}^j = -\Omega_{p0}/\Omega_c$ is proportional to the Rabi frequency of the initial probe field Ω_{p0} [25,28,30,31,44–46].

During the propagation of the slow light pulse the probe and control beams drive the jth atom to the dark state [28]

$$|\Psi^j\rangle = A\left(|g^j\rangle - \frac{\Omega_{p0}}{\Omega_c}|s^j\rangle\right) = A\left(1 - \frac{\Omega_{p0}}{\Omega_c}\sigma_{sg}^j\right)|g^j\rangle, \quad (5)$$

where

$$A = \left(1 + \Omega_{p0}^2/\Omega_c^2\right)^{-1/2} \quad (6)$$

is a normalization factor. Initially atoms are uncorrelated, thus the full quantum state of the atomic ensemble is

$$|\Psi\rangle = \prod_j |\Psi^j\rangle = A^N \prod_j \left(1 - \frac{\Omega_{p0}}{\Omega_c}\sigma_{sg}^j\right)|\mathbf{g}\rangle, \quad (7)$$

where N is a total number of atoms in the sample and $|\mathbf{g}\rangle = \prod_j |g^j\rangle$ is a complete atomic ground state. Note, that one can rewrite Eq. (7) in terms of spin-wave excitations:

$$|\Psi\rangle = A^N|\mathbf{g}\rangle + A^N \sum_{n=1}^{N} \left(-\frac{\Omega_{p0}}{\Omega_c}\right)^n |\Psi_n\rangle, \quad (8)$$

where

$$|\Psi_n\rangle = \sum_{j_1,\ldots,j_n} \sigma_{sg}^{j_1} \ldots \sigma_{sg}^{j_n}|\mathbf{g}\rangle \quad (9)$$

describes the state with n spin excitations. Since the incident probe field Ω_p represents a pulse of a classical light, the full state of the atoms $|\Psi\rangle$ is separable.

By switching off the control field, the probe field is stored in the atomic coherences ρ_{sg}^j [25–27]. A quantum state of the atomic ensemble is then given by Eq. (7). Immediately after switching off the control laser, the $\pi/2$ optical pulse is applied that couples the Rydberg s and p states and converts the Rydberg s state to a superposition of the s and p Rydberg states $|+\rangle = (|s\rangle + |p\rangle)/\sqrt{2}$, as shown in Fig. 1(b). Consequently, the state vector of the stored dark-state polariton is converted to

$$|\Psi_+\rangle = A^N \prod_j \left(1 - \frac{\Omega_{p0}}{\Omega_c}\sigma_{+g}^j\right)|\mathbf{g}\rangle. \quad (10)$$

Here the symmetric an antisymmetric creation operators are defined as

$$\sigma_{\pm g}^j = \frac{1}{\sqrt{2}}\left(\sigma_{sg}^j \pm \sigma_{pg}^j\right). \quad (11)$$

Note that the inverse transform reads

$$\sigma_{sg}^j = \frac{1}{\sqrt{2}}\left(\sigma_{+g}^j + \sigma_{-g}^j\right), \quad (12)$$

$$\sigma_{pg}^j = \frac{1}{\sqrt{2}}\left(\sigma_{+g}^j - \sigma_{-g}^j\right). \quad (13)$$

Calling on Eq. (11), it is convenient to represent the initial state given by Eq. (10) in terms of the bare atomic states

$$|\Psi_+\rangle = A^N \prod_j \left(1 - \frac{\Omega_{p0}}{\sqrt{2}\Omega_c}\left(\sigma_{sg}^j + \sigma_{pg}^j\right)\right)|\mathbf{g}\rangle. \quad (14)$$

B. Atomic state affected by the atom-atom interaction

During the storage the atoms undergo a free evolution without influence of the optical fields, yet affected by the atom-atom interaction $\mathcal{H}_{at-at}$ given by Eq. (4). During such an evolution the atomic state vector $|\Psi_+\rangle$ given by Eq. (10) transforms to

$$|\Psi(T)\rangle = e^{-i\mathcal{H}_{at-at}T}|\Psi_+\rangle, \quad (15)$$

where T is the storage time. By collecting the terms containing double sums as is detailed in Appendix A and using Eq. (10), the action of the evolution operator on the atomic state can be written as

$$|\Psi(T)\rangle = |\Psi_+\rangle + \sum_{j\neq j'}[e^{-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T} - 1]\sigma_{ps}^j\sigma_{sp}^{j'}|\Psi_+\rangle + \text{nonpair terms}. \quad (16)$$

The terms that are not written explicitly in Eq. (16) contain triple and higher sums. In Eq. (16) we used the fact that the operators σ_{sg}^j and σ_{pg}^j enter symmetrically the initial state vector $|\Psi_+\rangle$ given by Eq. (14). Consequently, the action of the operator $\sigma_{ps}^j\sigma_{sp}^{j'}$ on the initial state vector $|\Psi_+\rangle$ gives the same result as the action of the operator $\sigma_{pp}^j\sigma_{ss}^{j'}$ on $|\Psi_+\rangle$. This allows us to combine $\cos[V(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]$ and $-i\sin[V(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]$ entering Eq. (A10) into a single exponential function when acting on the initial state vector $|\Psi_+\rangle$.

From now on we will omit the terms due to the interaction involving three and more atoms. Such an assumption is legitimate if the density of Rydberg atoms is small enough and the duration of the storage time T is sufficiently short, so that it is unlikely to have more than a single pair of strongly interacting close-by Rydberg atoms. This is the case if a characteristic distance r_c, at which the RDDI potential $V(r_c)$ becomes equal to the inverse storage time T^{-1}, is smaller than a mean distance $r_{Ry} = n_{Ry}^{-1/3}$ between the atoms excited to the Rydberg state, i.e., $r_c \lesssim r_{Ry}$. Here

$$n_{Ry} = A^2\frac{\Omega_{p0}^2}{\Omega_c^2}n \quad (17)$$

is a density of Rydberg atoms, n is a total density of atoms, and $\Omega_{p0}^2/\Omega_c^2 \ll 1$ is a probability for an individual atom to be

excited to the Rydberg s state during the initial propagation of slow light. The RDDI potential depends on the distance between atoms as $V(r) = C_3/r^3$, giving a characteristic distance

$$r_c = (C_3 T)^{1/3}. \tag{18}$$

The condition $r_c \lesssim r_{Ry}$ then leads to an upper limit for the storage time $T_{max} = (C_3 n_{Ry})^{-1}$. A more detailed discussion of the validity is given in Appendix B.

In writing Eq. (16) we have included multiple RDDI transitions within the same pair of atoms providing an oscillating term $\exp[-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]$ which regularizes a divergent behavior of individual terms in the expansion of the evolution operator in the powers of T. This is important at interatomic distances $|\mathbf{r}_j - \mathbf{r}_{j'}| < r_c$ for which the RDDI energy $V(|\mathbf{r}_j - \mathbf{r}_{j'}|)$ exceeds T^{-1}.

Taking into account Eq. (14) for the initial state vector $|\Psi_+\rangle$, Eq. (16) yields the following state vector of the atomic system at the end of the free evolution:

$$|\Psi(T)\rangle \approx |\Psi_+\rangle + A^N \frac{\Omega_{p0}^2}{2\Omega_c^2} \sum_{j\neq j'} [e^{-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T} - 1]\sigma_{sg}^j \sigma_{pg}^{j'} \times \prod_{j''\neq j,j'} \left(1 - \frac{\Omega_{p0}}{\Omega_c}\sigma_{+g}^{j''}\right)|\mathbf{g}\rangle. \tag{19}$$

Using Eqs. (12) and (13), a pair of operators $\sigma_{sg}^j \sigma_{pg}^{j'}$ entering the above equation can be cast in terms of the operators $\sigma_{\pm g}^j$ and $\sigma_{\pm g}^{j'}$ as

$$\sigma_{sg}^j \sigma_{pg}^{j'} = \left(\sigma_{+g}^j + \sigma_{-g}^j\right)\left(\sigma_{+g}^{j'} - \sigma_{-g}^{j'}\right)/2. \tag{20}$$

Since the summation indices can be interchanged, the mixed terms containing the operators $-\sigma_{+g}^j \sigma_{-g}^{j'}$ and $\sigma_{-g}^j \sigma_{+g}^{j'}$ cancel each other in Eq. (19), so $\sigma_{sg}^j \sigma_{pg}^{j'}$ can be replaced by $(\sigma_{+g}^j \sigma_{+g}^{j'} - \sigma_{-g}^j \sigma_{-g}^{j'})/2$, giving

$$|\Psi(T)\rangle \approx |\Psi_+\rangle + A^N \frac{\Omega_{p0}^2}{4\Omega_c^2} \sum_{j\neq j'} [e^{-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T} - 1] \times \left(\sigma_{+g}^j \sigma_{+g}^{j'} - \sigma_{-g}^j \sigma_{-g}^{j'}\right) \prod_{j''\neq j,j'} \left(1 - \frac{\Omega_{p0}}{\Omega_c}\sigma_{+g}^{j''}\right)|\mathbf{g}\rangle. \tag{21}$$

The states of the atoms at the end of the free evolution are schematically shown in Fig. 2.

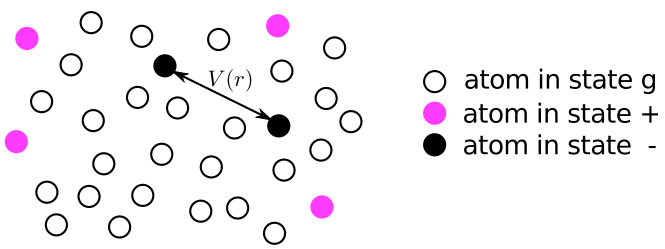

FIG. 2. Schematic depiction of the state of atoms at the end of storage period.

Just before the retrieval one applies another $\pi/2$ optical pulse that couples the Rydberg states s and p. This converts the state $|+\rangle = (|s\rangle + |p\rangle)/\sqrt{2}$ into the Rydberg state $|p\rangle$, whereas the state $|-\rangle = (|s\rangle - |p\rangle)/\sqrt{2}$ is converted back into the Rydberg state $|s\rangle$ [see Fig. 1(b)]. As a result the state vector (21) reduces to

$$|\Psi_{fin}\rangle \approx |\Psi_p\rangle + A^N \frac{\Omega_{p0}^2}{4\Omega_c^2} \sum_{j\neq j'} [e^{-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T} - 1] \times \left(\sigma_{pg}^j \sigma_{pg}^{j'} - \sigma_{sg}^j \sigma_{sg}^{j'}\right) \prod_{j''\neq j,j'} \left(1 - \frac{\Omega_{p0}}{\Omega_c}\sigma_{pg}^{j''}\right)|\mathbf{g}\rangle, \tag{22}$$

where $|\Psi_p\rangle$ is obtained from Eq. (10) with σ_{+g}^j replaced by σ_{pg}^j:

$$|\Psi_p\rangle = A^N \prod_j \left(1 - \frac{\Omega_{p0}}{\Omega_c}\sigma_{pg}^j\right)|\mathbf{g}\rangle. \tag{23}$$

The second term in Eq. (22) represents the correlated pairs of atoms in the Rydberg s and p states created due to the atom-atom interaction. As we shall see in Sec. IV, the Rydberg s excitations are converted into pairs of correlated photons during the restoring of the slow light. The first term in Eq. (22) does not contain the Rydberg s state and thus will not contribute to the restored slow light.

C. Atomic correlation functions

The spectral density of the restored light is related to the atomic first-order correlation at different sites j and j',

$$G_{at}^{(1)}(\mathbf{r}_j, \mathbf{r}_{j'}) = \langle \Psi_{fin} | \sigma_{gs}^{j\dagger} \sigma_{gs}^{j'} | \Psi_{fin} \rangle. \tag{24}$$

Using the approximate expression (22) for the final state we get

$$G_{at}^{(1)}(\mathbf{r}_j, \mathbf{r}_{j'}) \approx A^4 \frac{\Omega_{p0}^4}{4\Omega_c^4} \sum_{j''\neq j,j'} [e^{iV(|\mathbf{r}_j - \mathbf{r}_{j''}|)T} - 1] \times [e^{-iV(|\mathbf{r}_{j'} - \mathbf{r}_{j''}|)T} - 1]. \tag{25}$$

The intensity of the restored light is related to the density of atoms in the Rydberg state $|s\rangle$. The probability for an atom to be in this state can be obtained from the atomic correlation function at the same site j:

$$G_{at}^{(1)}(\mathbf{r}_j) \equiv G_{at}^{(1)}(\mathbf{r}_j, \mathbf{r}_j) = \langle \Psi_{fin} | \sigma_{gs}^{j\dagger} \sigma_{gs}^{j} | \Psi_{fin} \rangle. \tag{26}$$

Using Eq. (25) this probability reduces to

$$G_{at}^{(1)}(\mathbf{r}_j) \approx A^4 \frac{\Omega_{p0}^4}{2\Omega_c^4} \sum_{j'\neq j} (1 - \cos[V(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]). \tag{27}$$

The atomic second-order correlation function

$$G_{at}^{(2)}(\mathbf{r}_j, \mathbf{r}_{j'}) = \langle \Psi_{fin} | \sigma_{gs}^{j\dagger} \sigma_{gs}^{j'\dagger} \sigma_{gs}^{j'} \sigma_{gs}^{j} | \Psi_{fin} \rangle \tag{28}$$

using Eq. (22) for $|\Psi_{fin}\rangle$ can be expressed as

$$G_{at}^{(2)}(\mathbf{r}_j, \mathbf{r}_{j'}) \approx (1 - \delta_{j,j'}) A^4 \frac{\Omega_{p0}^4}{2\Omega_c^4} \{1 - \cos[V(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]\}. \tag{29}$$

The atomic second-order correlation function (29) is related to the second-order correlation function of the restored light $G^{(2)}(\tau)$ via Eq. (32) presented below.

IV. PROPERTIES OF RESTORED PROBE PULSE

After application of the second $\pi/2$ pulse the probe pulse of light is restored by switching on the control beam characterized by the Rabi frequency Ω_c. The state of the atomic ensemble just before the retrieval is given by Eq. (22). During the retrieval only the atoms in the Rydberg state s contribute to the probe beam. The p state excitations remain in the medium and thus will no longer be considered [47]. The restored probe field

$$\Omega_p(\mathbf{r}_j) = -\Omega_c \sigma^j_{gs}(T) \tag{30}$$

is generated from atomic coherences $\sigma^j_{gs}(T)$ involving the Rydberg s state.

Note that the light restored from the atomic state (22) consist of pairs of correlated photons corresponding to the second term in Eq. (22), there being no contributions by single photons. Single photons can appear only due to losses, when one of the photons forming the pair is absorbed. Yet, as we will see later in this section, the absorption does not significantly distort the second-order correlation function for a sufficiently large separation between the photons.

The second term of Eq. (22) describes a superposition of all possible atomic pairs, so the restored light is in a quantum superposition of all corresponding photon pairs. The measurement of the second-order correlation function of the restored light selects a photon pair in which the photons are separated by a chosen distance.

A. Second-order correlation function of the restored light

The second-order correlation function of the retrieved light

$$G^{(2)}(\tau) = \langle \Omega_p^\dagger(t)\Omega_p^\dagger(t+\tau)\Omega_p(t+\tau)\Omega_p(t)\rangle \tag{31}$$

is calculated by averaging the atomic second-order correlation function $G^{(2)}_{at}(\mathbf{r}_j,\mathbf{r}_{j'}) = \langle \Psi_{fin}|\sigma^{j\dagger}_{gs}\sigma^{j'\dagger}_{gs}\sigma^{j'}_{gs}\sigma^{j}_{gs}|\Psi_{fin}\rangle$ over the atomic positions $\mathbf{r}_j$ and $\mathbf{r}_{j'}$ separated by $|z_j - z_{j'}| = v_{g0}\tau$ along the propagation direction z. Thus the correlation function of the restored light reads

$$G^{(2)}(\tau) = \Omega_c^4/N_r \sum_{j,j'} G^{(2)}_{at}(\mathbf{r}_j,\mathbf{r}_{j'}), \tag{32}$$

where the summation extends over a narrow region, shown in Fig. 3, in which the atomic second-order correlation function is averaged. Here N_r is the number of atoms used in averaging. If the width of the atomic medium is smaller than the separation distance $v_{g0}\tau$, the averaging does not significantly alter the atomic second-order correlation function $G^{(2)}_{at}(\mathbf{r}_j,\mathbf{r}_{j'})$. By concentrating on the distances between the atoms $|\mathbf{r}-\mathbf{r}'|$ larger than the width of the atom cloud the problem becomes essentially one-dimensional.

From Eq. (29) follows that the second-order correlation function of the retrieved light is determined by the atom-atom interaction:

$$G^{(2)}(\tau) \propto 1 - \cos[V(v_{g0}\tau)T]. \tag{33}$$

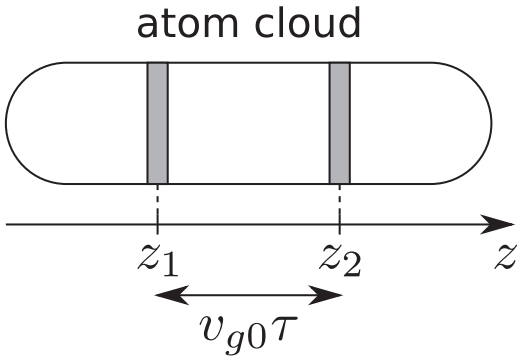

FIG. 3. Regions (gray color) over which the atomic second-order correlation function $G^{(2)}_{at}$ is averaged in calculating the second-order correlation function of the restored light.

One can see that the scale of the distances $v_{g0}\tau$ probed by the second-order correlation function depends on the storage time T. For a sufficiently small storing time T and a large delay time τ, $V(v_{g0}\tau)T \ll \pi$, the second-order correlation function of the retrieved light is proportional to the square of the interaction potential at the interatomic distance $v_{g0}\tau$, i.e., $G^{(2)}(\tau) \propto [V(v_{g0}\tau)T]^2$. In this way the restored pulse is created exclusively due to the atom-atom interaction which vanishes as interatomic distance increases. The probability to find in the restored pulse a pair of photons separated by large distances goes to zero, and the (unnormalized) second-order correlation function $G^{(2)}(\tau)$ decays as τ increases.

B. Estimation of the intensity of the restored light

Let us estimate the intensity of the restored probe pulse. The restored field is generated from atomic coherences $\sigma^j_{gs}(T)$ involving the Rydberg s state, according to the equation $\Omega_p(\mathbf{r}_j) = -\Omega_c\sigma^j_{gs}(T)$. Thus the intensity, proportional to $|\Omega_p|^2$, can be calculated using atomic first-order correlation function $G^{(1)}_{at}$. Calling on Eq. (27) the ratio of the intensities of the restored and the incoming probe pulses is given by

$$\frac{\Omega^2_{p,out}}{\Omega^2_{p0}} = A^4 \frac{\Omega^2_{p0}}{2\Omega^2_c} \sum_j \{1 - \cos[V(|\mathbf{r}-\mathbf{r}_j|)T]\}. \tag{34}$$

The sum in Eq. (34) can be estimated as

$$\sum_j \{1 - \cos[V(|\mathbf{r}-\mathbf{r}_j|)T]\} \approx n \int d\mathbf{r}' \{1 - \cos[V(|\mathbf{r}-\mathbf{r}'|)T]\} \sim n r_c^3, \tag{35}$$

where we have used Eq. (B4) for evaluating the integral. Using Eq. (17) for the density of Rydberg atoms, we obtain that the ratio of the intensities of the restored and the incoming probe pulses is of the order of

$$\frac{\Omega^2_{p,out}}{\Omega^2_{p0}} \sim n_{Ry} r_c^3. \tag{36}$$

Note that in order to neglect the interaction involving three or more Rydberg atoms we require that $n_{Ry} r_c^3 \ll 1$. Thus the intensity of the restored light is much smaller than that of the

incident light. Consequently most of the probe pulse remains in the medium in the form of excitations of the Rydberg p state.

C. Spectral width of the restored light

The spectrum $S(\omega)$ is related to the first-order correlation function of the light

$$G^{(1)}(\tau) = \langle \Omega_{\rm p}^{\dagger}(t+\tau)\Omega_{\rm p}(t)\rangle \tag{37}$$

via the equation

$$S(\omega) = \int e^{-i\omega\tau} G^{(1)}(\tau) d\tau. \tag{38}$$

The restored probe field $\Omega_{\rm p}(\mathbf{r}_j) = -\Omega_{\rm c}\sigma_{gs}^{j}(T)$ is generated from atomic coherences $\sigma_{gs}^{j}(T)$ involving the Rydberg s state. Therefore the spectral width can be calculated from the atomic first-order correlation function $G_{\rm at}^{(1)}$ at different sites. Using Eq. (25) we get

$$G^{(1)}(\tau) \sim \sum_j [e^{iV(|\mathbf{r}-\mathbf{r}_j|)T} - 1][e^{-iV(|\mathbf{r}-\mathbf{r}_j+v_{g0}\tau\hat{\mathbf{e}}_z|)T} - 1]. \tag{39}$$

We see that the restored light acquires a finite width of the spectrum, even when the incident probe beam is monochromatic. From Eq. (39) one can estimate the characteristic width of the function $G^{(1)}(\tau)$ to be of the order of $r_{\rm c}/v_{g0}$, where $r_{\rm c}$ is defined as $V(r)T = (r_{\rm c}/r)^3$. Thus the spectral width of the restored light $S(\omega)$ is of the order of $v_{g0}/r_{\rm c}$.

D. Influence of losses for the two-photon correlation measurements

Since the restored light has a finite spectral width $v_{g0}/r_{\rm c}$, it experiences losses due to a finite transmittivity width of the EIT window. To estimate the influence of losses occurring during the propagation of the restored light in the atomic medium, let us consider a part of the atomic state containing only pairs of atoms in the Rydberg s state. According to Eq. (22), this part is

$$|\Psi_{\rm ss}\rangle = \frac{\Omega_{\rm p0}^2}{4\Omega_{\rm c}^2}\sum_{j\neq j'}[e^{-iV(|\mathbf{r}_j-\mathbf{r}_{j'}|)T} - 1]\sigma_{gs}^{j'\dagger}\sigma_{gs}^{j\dagger}|\mathbf{g}\rangle. \tag{40}$$

We are interested in the delay times τ entering the second-order photon correlation function $G^{(2)}(\tau) = \langle \Omega_{\rm p}^{\dagger}(t)\Omega_{\rm p}^{\dagger}(t+\tau)\Omega_{\rm p}(t+\tau)\Omega_{\rm p}(t)\rangle$, such that $v_{g0}\tau$ is larger than the width of the atom cloud. Such delay times correspond to the distances between atoms $|\mathbf{r}-\mathbf{r}'|$ larger than the width of the medium. In this case we can consider the state $|\Psi_{\rm ss}\rangle$ written in terms of the spin-flip operators averaged over the cross section of the medium:

$$|\Psi_{ss}\rangle = C\int dz \int dz' I(|z-z'|)\sigma_{gs}^{\dagger}(z')\sigma_{gs}^{\dagger}(z)|\mathbf{g}\rangle, \tag{41}$$

where

$$I(z) \equiv i(e^{-iV(z)T} - 1), \qquad C = \frac{\Omega_{\rm p0}^2}{4\Omega_{\rm c}^2}n^2S^2. \tag{42}$$

Introducing the spin-flip operators in the momentum representation

$$\sigma_{gs,k} = \frac{1}{\sqrt{2\pi}}\int dz\, e^{ikz}\sigma_{gs}(z) \tag{43}$$

and using Eq. (41) we get

$$|\Psi_{ss}\rangle = \frac{C}{2\pi}\int dk \int dk' \int dz \int dz' I(|z-z'|)e^{ikz+ik'z'} \times \sigma_{gs,k}^{\dagger}\sigma_{gs,k'}^{\dagger}|\mathbf{g}\rangle. \tag{44}$$

By separating the mean momentum $\bar{k} = \frac{1}{2}(k+k')$ and the difference $\Delta k = \frac{1}{2}(k-k')$, the atomic state $|\Psi_{ss}\rangle$ can be written as

$$|\Psi_{ss}\rangle = \frac{C}{2\pi}\int d\Delta k\, \tilde{I}(\Delta k)\sigma_{gs,\Delta k}^{\dagger}\sigma_{gs,-\Delta k}^{\dagger}|\mathbf{g}\rangle, \tag{45}$$

where

$$\tilde{I}(k) = i\int dz\,[e^{-iV(|z|)T} - 1]e^{ikz} \tag{46}$$

is the Fourier transform of $I(z)$. After restoring the light, the spin-flip operators $\sigma_{gs,k}$ are replaced by operators for dark-state polaritons $\mathcal{P}_{{\rm D},k} = \frac{v_{g0}}{c}\frac{\Omega_{{\rm p},k}}{\Omega_{\rm c}} - \sigma_{gs,k}$, giving the state

$$|\Phi\rangle = \frac{C}{2\pi}\int dk\, \tilde{I}(k)\mathcal{P}_{{\rm D},k}^{\dagger}\mathcal{P}_{{\rm D},-k}^{\dagger}|{\rm vac}\rangle. \tag{47}$$

This state describes a pair of polaritons with the wave vectors k and $-k$ around the zero central wave vector: $\bar{k} = 0$.

The propagation duration of the restored polariton is of the order of

$$\tau_{\rm prop} \sim \frac{L}{2v_{g0}}, \tag{48}$$

assuming that it propagates approximately half of a medium length L. During the propagation the dark-state polaritons decay due to nonadiabatic losses with the rate $\gamma_{\rm pol} = 2\Gamma(v_{g0}k)^2/\Omega_{\rm c}^2$ [29]. Thus the polariton operator $\mathcal{P}_{{\rm D},k}$ changes to

$$\mathcal{P}_{{\rm D},k}e^{-(L^2/2\alpha)k^2} + \eta_k b_k\,, \quad \text{where} \quad \eta_k = \sqrt{1-e^{-(L^2/\alpha)k^2}}. \tag{49}$$

The exponent $L^2k^2/(2\alpha) = \gamma_{\rm pol}\tau_{\rm prop}$ describes the polariton decay. Additional bosonic noise operators b_k have been included to preserve the commutation relations [43]. The final state then becomes

$$|\Phi'\rangle \sim \int dk\, \tilde{I}(k)(\mathcal{P}_{{\rm D},k}^{\dagger}e^{-(L^2/2\alpha)k^2} + \eta_k b_k^{\dagger}) \times (\mathcal{P}_{{\rm D},-k}^{\dagger}e^{-(L^2/2\alpha)k^2} + \eta_k b_{-k}^{\dagger})|{\rm vac}\rangle. \tag{50}$$

We are interested in the second-order correlation function

$$G^{(2)}(z,z') = \langle\Phi'|\mathcal{P}_{\rm D}^{\dagger}(z)\mathcal{P}_{\rm D}^{\dagger}(z')\mathcal{P}_{\rm D}(z')\mathcal{P}_{\rm D}(z)|\Phi'\rangle. \tag{51}$$

It is noteworthy that only the component of the state vector $|\Phi'\rangle$ containing two polariton operators,

$$\int dk\, \tilde{I}(k)\mathcal{P}_{{\rm D},k}^{\dagger}\mathcal{P}_{{\rm D},-k}^{\dagger}e^{-(L^2/\alpha)k^2}|{\rm vac}\rangle, \tag{52}$$

contributes to $G^{(2)}(z,z')$. Therefore, going back to the coordinate representation we obtain

$$\int dz \int dz'\, I'(z-z')\mathcal{P}_{\rm D}^{\dagger}(z)\mathcal{P}_{\rm D}^{\dagger}(z')|{\rm vac}\rangle, \tag{53}$$

where

$$I'(z) \approx \frac{1}{2\pi}\int dk\, \tilde{I}(k)e^{-ikz-(L^2/\alpha)k^2} = i\frac{\sqrt{\alpha}}{2L\sqrt{\pi}}\int d\tilde{z}\,[e^{-iV(|\tilde{z}|)T}-1]e^{-(\alpha/4L^2)(\tilde{z}-z)^2}. \tag{54}$$

We see that losses modify the coefficient $e^{-iV(z)T}-1$ appearing in Eq. (22) by convolving it with a Gaussian $\exp[-\alpha(z/2L)^2]$. Combining Eqs. (51) and (53), we obtain that the polariton losses change the second-order correlation function $|I(z-z')|^2$ to $G^{(2)}(z,z')=|I'(z-z')|^2$. Consequently the second-order correlation function of the restored light is determined by a modified potential influenced by the losses, rather than by an actual interaction potential V.

The second-order correlation function with and without inclusion of losses is displayed in Fig. 4. In order to get a dimensionless quantity, the second-order correlation function shown in Fig. 4 is normalized using the intensity of the initial probe pulse. As the dashed red curve in Fig. 4 indicates, the losses during the propagation of light lead to the smoothening of the second-order correlation function, in agreement with Eq. (54). One can see that the the influence of losses diminishes for distances $z = v_{g0}\tau$ much larger than a characteristic loss distance $L/\sqrt{\alpha}$. This corresponds to a delay time τ greater than $\Gamma\sqrt{\alpha}/\Omega_{\rm c}^2$. For smaller delay times τ the effects of the losses become significant, reducing the number of photon pairs.

Note that the spectral width of the restored light, considered in the Sec. IV C, is related to the first-order correlation function rather than to the second-order correlation function shown in Fig. 4. In general those two correlation functions are not directly related; only for chaotic classical light sources is the second-order correlation $g^{(2)}$ determined by the first-order correlation $g^{(1)}$ [42,43]. Thus, in the absence of losses the spectral width does not limit the structure of the second-order correlation function. This is not the case when losses are present, as one can see from the red dashed curve in Fig. 4.

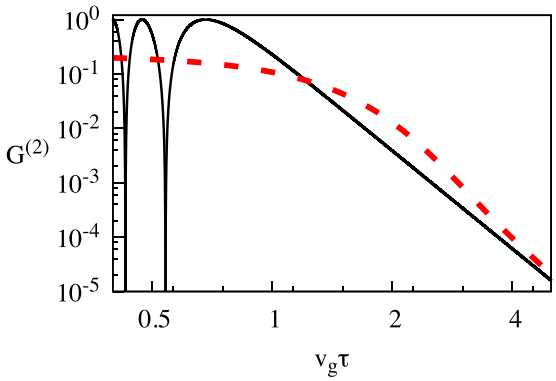

FIG. 4. The second-order correlation function of the restored light normalized to the intensity of the input pulse without including losses (solid black line) and modified by polariton losses (dashed red line). The distances are measured in units of $r_{\rm c}$, the characteristic distance of losses is assumed to be $L/\sqrt{\alpha} = 1/2$.

V. DISCUSSION AND CONCLUSIONS

The proposed ladder scheme can be experimentally implemented using ultracold ^{87}Rb atoms [11] by preparing the atoms in a hyperfine ground state $|5S_{1/2}, F=2\rangle$ serving as the state $|g\rangle$ in our scheme. A hyperfine excited state $|5P_{3/2}, F=3\rangle$ with a decay rate $\Gamma = 2\pi \times 6$ MHz corresponds to the state $|e\rangle$. The characteristic distance $r_{\rm c}$ of the order $r_{\rm c} \approx 0.18$ mm can be achieved for the storage duration T of the order of 10 μs and a principal quantum number of the Rydberg levels $n = 100$, the latter leading to the coefficient C_3 of the order of 610 GHz μm^3. The interaction potential $V(r_{\rm c}) = 0.1$ MHz can be much smaller than the microwave Rabi frequency, which has been created of the order of 100 MHz [13,48]. Note that for such a large principal quantum number a strong Rydberg blockade occurs, with the blockade radius of the order of 13 μm [11]. Yet here we are spectroscopically probing the interatomic distances larger than the Rydberg blockade radius, so the blockade effects are not important. On the other hand, the polariton losses due to the finite spectral width of the regenerated light can be neglected for distances between the emitting atomic pairs $v_{g0}\tau$ larger than 0.18 mm when using the experimentally accessible length of the atomic medium $L = 1$ mm and the optical density $\alpha = 30$ [49,50]. The Rabi frequency of the control beam $\Omega_{\rm c} = 2\pi \times 2$ MHz leads to the group velocity of the polaritons $v_{g0} = 140$ m/s, thus these distances correspond to the delay time $\tau \approx 1.3$ μs. In this way, it is feasible to observe correlated photon pairs produced by storing and regenerating the Rydberg slow light.

The proposed method can be applied not only to the resonant dipole-dipole interactions but also to the other types of atom-atom interactions. The suggested Ramsey-type scheme can be employed to generate narrow-linewidth biphotons with correlation times of the order of the propagation delay time. The scheme can also provide an efficient way for manipulation of individual photons or operation of qubits, since two photons can interact with each other effectively at relatively large distances determined by the interaction between the Rydberg atoms. With increasing the storage time (e.g., by using an optical lattice to confine the atomic motion within a distance smaller than the wavelength [51]), the scheme may be used as a sensitive tool for probing the atom-atom interaction. In this way our proposal offers possibilities and applications in generation of nonclassical light, manipulation of quantum information, and precision measurement of long-distance interaction.

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APPENDIX A: INTERACTION OF A PAIR OF ATOMS

Let us consider the free evolution operator $e^{-i\mathcal{H}_{\rm at-at}T}$. For further approximations it is convenient to represent the operator $e^{-i\mathcal{H}_{\rm at-at}T}$ as

$$e^{-i\mathcal{H}_{\rm at-at}T} = 1 + (e^{-i\mathcal{H}_{\rm at-at}T} - 1). \tag{A1}$$

Since the duration of the evolution T is considered to be short enough, the first term (the unit operator not changing the state) represents a dominant contribution to the evolution, whereas the remaining term in Eq. (A1) takes care of the changes in the state vector due to the atom-atom interaction. Because of the short storage time, the latter term mostly couples a pair of atoms. Expanding the exponent $e^{-i\mathcal{H}_{\rm at-at}T}$ into Taylor series we have

$$e^{-i\mathcal{H}_{\rm at-at}T} - 1 = -iT\mathcal{H}_{\rm at-at} + \tfrac{1}{2}(-iT)^2\mathcal{H}^2_{\rm at-at} + \cdots . \tag{A2}$$

Calling on Eq. (3) of the main text for the interaction Hamiltonian $\mathcal{H}_{\rm at-at}$, the first term in the Taylor expansion (A2) reads

$$-iT\mathcal{H}_{\rm at-at} = -i\sum_{j\neq j'} V(|\mathbf{r}_j - \mathbf{r}_{j'}|)T\sigma^j_{ps}\sigma^{j'}_{sp}. \tag{A3}$$

The term appearing in the second order of the expansion (A2) is

$$(-iT)^2\mathcal{H}^2_{\rm at-at} = \sum_{j\neq j'}\sum_{j''\neq j'''} V(|\mathbf{r}_j - \mathbf{r}_{j'}|)V(|\mathbf{r}_{j'''} - \mathbf{r}_{j''}|) \times \sigma^j_{ps}\sigma^{j'}_{sp}\sigma^{j''}_{ps}\sigma^{j'''}_{sp}. \tag{A4}$$

When $j = j''$, $j' = j'''$ or $j = j'''$, $j' = j''$, the summation in this expression runs only over two indices. Since $\sigma^j_{ps}\sigma^j_{ps} = 0$, the second-order term can be separated into two parts, the first part containing double summation, the second part containing higher sums,

$$(-iT)^2\mathcal{H}^2_{\rm at-at} = \sum_{j\neq j'}[-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]^2\sigma^j_{pp}\sigma^{j'}_{ss} + \text{nonpair terms}. \tag{A5}$$

In a similar manner, the third-order term can be represented as

$$(-iT)^3\mathcal{H}^3_{\rm at-at} = \sum_{j\neq j'}[-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]^3\sigma^j_{ps}\sigma^{j'}_{sp} + \text{nonpair terms}. \tag{A6}$$

In this way, the pair summation in the cubic term contains the same operators $\sigma^j_{ps}\sigma^{j'}_{sp}$ as the first-order term. Repeating the same procedure one arrives at the following general result for the odd and even terms in the expansion (A2):

$$(-iT)^{2m+1}\mathcal{H}^{2m+1}_{\rm at-at} = \sum_{j\neq j'}[-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]^{2m+1}\sigma^j_{ps}\sigma^{j'}_{sp} + \text{nonpair terms}, \tag{A7}$$

$$(-iT)^{2m}\mathcal{H}^{2m}_{\rm at-at} = \sum_{j\neq j'}[-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]^{2n}\sigma^j_{pp}\sigma^{j'}_{ss} + \text{nonpair terms}, \tag{A8}$$

with $m = 0, 1, \ldots$. Thus, collecting in each power of the Hamiltonian $\mathcal{H}_{\rm at-at}$ only the terms containing double sums, Eq. (A1) becomes

$$e^{-i\mathcal{H}_{\rm at-at}T} = 1 + \sum_{m=0}^{\infty}\frac{1}{(2m+1)!} \times \sum_{j\neq j'}[-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]^{2m+1}\sigma^j_{ps}\sigma^{j'}_{sp} + \sum_{m=1}^{\infty}\frac{1}{(2m)!}\sum_{j\neq j'}[-iV(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]^{2m}\sigma^j_{pp}\sigma^{j'}_{ss} + \text{nonpair terms}, \tag{A9}$$

where the terms that are not written explicitly contain triple and higher sums. After summation we obtain

$$e^{-i\mathcal{H}_{\rm at-at}T} - 1 = \sum_{j\neq j'}\{\cos[V(|\mathbf{r}_j - \mathbf{r}_{j'}|)T] - 1\}\sigma^j_{pp}\sigma^{j'}_{ss} - i\sum_{j\neq j'}\sin[V(|\mathbf{r}_j - \mathbf{r}_{j'}|)T]\sigma^j_{ps}\sigma^{j'}_{sp} + \text{nonpair terms}. \tag{A10}$$

APPENDIX B: VALIDITY OF THE APPROXIMATION

Let us consider the necessary conditions when the approximate expression (21) for the atomic state is valid. The conditions can be obtained by requiring the state $|\Psi(T)\rangle$ to be normalized, that is $\langle\Psi(T)|\Psi(T)\rangle \approx 1$.

Equation (21) can be separated into two parts, $|\Psi(T)\rangle = |\Psi_+\rangle + |\Delta\Psi\rangle$. Since the initial state is normalized, $\langle\Psi_+|\Psi_+\rangle = 1$, the normalization condition for the final state $|\Psi(T)\rangle$ reads

$$2\,\mathrm{Re}\langle\Psi_+|\Delta\Psi\rangle + \langle\Delta\Psi|\Delta\Psi\rangle = 0. \tag{B1}$$

Using Eq. (21) we obtain

$$\begin{aligned}2\,\mathrm{Re}&\langle\Psi_+|\Delta\Psi\rangle + \langle\Delta\Psi|\Delta\Psi\rangle \\ &\approx A^6\frac{\Omega^6_{\rm p0}}{4\Omega^6_{\rm c}}\sum_{j\neq j'\neq j''}[e^{iV(|\mathbf{r}_j-\mathbf{r}_{j'}|)T} - 1][e^{-iV(|\mathbf{r}_j-\mathbf{r}_{j''}|)T} - 1] \\ &\quad + A^8\frac{\Omega^8_{\rm p0}}{16\Omega^8_{\rm c}}\sum_{j\neq j'\neq j''\neq j'''}[e^{iV(|\mathbf{r}_j-\mathbf{r}_{j_r}|)T} - 1] \\ &\quad \times [e^{-iV(|\mathbf{r}_{j''}-\mathbf{r}_{j'''}|)T} - 1].\end{aligned} \tag{B2}$$

We can estimate the expressions in Eq. (B2) replacing summation by integration. Then we get

$$\begin{aligned}2\,\mathrm{Re}&\langle\Psi_+|\Delta\Psi\rangle + \langle\Delta\Psi|\Delta\Psi\rangle \\ &\approx A^6\frac{\Omega^6_{\rm p0}}{4\Omega^6_{\rm c}}n^3\int d\mathbf{r}\int d\mathbf{r}'[e^{iV(|\mathbf{r}-\mathbf{r}'|)T} - 1] \\ &\quad \times \int d\mathbf{r}''[e^{-iV(|\mathbf{r}-\mathbf{r}''|)T} - 1] \\ &\quad + A^8\frac{\Omega^8_{\rm p0}}{16\Omega^8_{\rm c}}n^4\int d\mathbf{r}\int d\mathbf{r}'[e^{iV(|\mathbf{r}-\mathbf{r}'|)T} - 1]\int d\mathbf{r}'' \\ &\quad \times \int d\mathbf{r}'''[e^{-iV(|\mathbf{r}''-\mathbf{r}'''|)T} - 1],\end{aligned} \tag{B3}$$

where n is the density of atoms. The integrals in Eq. (B3) can be estimated as follows. Using the interaction potential $V(r) = C_3/r^3$ we have

$$\int d\mathbf{r}'\{1-\cos[V(|\mathbf{r}-\mathbf{r}'|)T]\} = 4\pi\int_0^\infty r^2\{1-\cos[V(r)T]\}dr = \frac{2}{3}\pi^2 C_3 T \equiv \frac{2}{3}\pi^2 r_c^3, \quad \text{(B4)}$$

where $r_c = (C_3 T)^{1/3}$ is a characteristic distance at which the RDDI potential $V(r_c)$ becomes of the order of the inverse storage time T^{-1}. On the other hand, the integral

$$\int d\mathbf{r}' \sin[V(|\mathbf{r}-\mathbf{r}'|)T] = 4\pi\int_0^\infty r^2 \sin[V(r)T]dr$$

does not converge at large values of r. To get a finite value we should take into account a finite size of the atomic cloud. Then this integral becomes proportional to r_c^3. Thus the two terms in Eq. (B3) are of the order of $(n_{\mathrm{Ry}} r_c^3)^2 (n_{\mathrm{Ry}}\mathcal{V})$ and $(n_{\mathrm{Ry}} r_c^3)^2 (n_{\mathrm{Ry}}\mathcal{V})^2$, where $\mathcal{V}$ is the volume of the atomic cloud.

We can conclude that Eq. (B3) is close to zero and the approximation is valid when

$$n_{\mathrm{Ry}} r_c^3 \ll 1 \quad \text{(B5)}$$

and the total number $n_{\mathrm{Ry}}\mathcal{V}$ of Rydberg atoms in the atomic cloud is not large.

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