Single-photon superradiant beating from a Doppler-broadened ladder-type atomic ensemble

Yoon-Seok Lee,[1] Sang Min Lee,[1,2] Heonoh Kim,[1] and Han Seb Moon[1,*]

[1]*Department of Physics, Pusan National University, Geumjeong-Gu, Busan 46241, South Korea*

[2]*Currently with Korea Research Institute of Standards and Science, Daejeon 34113, South Korea*

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We report on heralded-single-photon superradiant beating in the spontaneous four-wave mixing process of Doppler-broadened ladder-type ^{87}Rb atoms. When Doppler-broadened atoms contribute to two-photon coherence, the detection probability amplitudes of the heralded single photons are coherently superposed despite inhomogeneous broadened atomic media. Single-photon superradiant beating is observed, which constitutes evidence for the coherent superposition of two-photon amplitudes from different velocity classes in the Doppler-broadened atomic ensemble. We present a theoretical model in which the single-photon superradiant beating originates from the interference between wavelength-separated two-photon amplitudes via the reabsorption filtering effect.

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Superradiance is a collective phenomenon whereby radiation is amplified by the coherence of multiple emitters, when identical atoms interact with a common vacuum mode. Since the concept of superradiance was first introduced by Dicke in 1954 [1], it has played prominent roles in the fields of optics, quantum mechanics, astrophysics, and quantum optics [2]. In particular, a single-photon superradiance (SPS) has attracted considerable interest from the quantum science community, including researchers investigating quantum communication, quantum computation, quantum repeaters, and quantum memory [3–6]. Thus far, SPS has usually been observed in cold atoms with high optical depth [7–11]. The dense, cold atomic media interact with a common vacuum mode, and their spatial phase coherence can be well preserved because the atoms are mostly motionless. Recently, SPS has been highlighted in various physical systems, such as quantum dots [12] and nitrogen-vacancy centers in diamond [13].

Superradiant beating in a Doppler-broadened atomic ensemble was first reported by the Haroche group in 1978 [14]. The occurrence of Doppler beats in superradiance is interesting, because it shows the collective effect of atoms moving with different velocities on light emission. The blue- and redshifted light signals emitted from two velocity groups moving in opposite directions are coherently superposed, and they interfere with each other even though they are emitted from physically different atoms. This interference effect has been demonstrated using classical light pulses in atomic vapor cell experiments with a four-wave mixing scheme, known as motion-induced signal revival [15]. This constructive interference of the collective excitation for Doppler-broadened atoms can be observed at a single-photon level. The observation of Doppler beats in superradiance is important because of the evidence of SPS in the Doppler-broadened atomic ensemble.

In this paper, we report on SPS beating between both two-photon amplitudes of the photon pairs generated from two separated atomic velocity groups in Doppler-broadened ladder-type ^{87}Rb warm vapor. Two-photon coherence, which corresponds to two-photon emission probability, is formed in the majority of velocity classes in our experimental scheme such that the probability amplitudes of detecting photons with difference frequencies are superposed. In this case, the temporal envelope of SPS can be characterized by the frequency distribution with respect to the atomic motions.

The experimental configuration is identical to that for the generation of correlated photon pairs from an atomic ensemble via spontaneous four-wave mixing [10,16], as shown in Fig. 1(a). The pump (Ω_P) and coupling (Ω_C) fields interact with the $5S_{1/2}$-$5P_{3/2}$ and $5P_{3/2}$-$5D_{5/2}$ transitions, respectively. The symbols δ_P and δ_C denote the detuning frequencies from the resonances of the pump and coupling fields, respectively, and the two-photon resonance condition is satisfied, i.e., $\delta_P + \delta_C = 0$. An atomic ensemble with a Maxwell-Boltzmann velocity distribution in a warm vapor can be coherently excited owing to the Doppler-free two-photon resonant configuration of Fig. 1(a).

The excited atoms spontaneously decay from the $5D_{5/2}$ state to the $5P_{3/2}$ state, emitting a signal photon. Upon heralding detection of the signal photon, the single atomic excitation in a collection of atoms in the intermediate state is conditionally prepared as

$$|\Psi\rangle = \frac{1}{\sqrt{N}} \sum_j C_j e^{i\delta_j t} e^{i\vec{k}_S \cdot \vec{r}_j} |g_1, \ldots, i_j, \ldots, g_N\rangle, \quad (1)$$

where N represents the number of atoms, $\vec{k}_S$ the wave vector of the signal field, δ_j the detuning of the incident field due to the Doppler shift, $\vec{r}_j$ the position of each atom in the ensemble, and C_j the probability amplitude of finding atoms in the intermediate state $|i\rangle$. In this N-atom entangled state essential for SPS, the so-called "Dicke-like state", a single atomic excitation is shared among a large number of moving atoms. Thus, the probability amplitudes of detecting an idler photon in the phase-matched direction are coherently superposed throughout the atomic spatial and velocity distributions. In Eq. (1), the spatial phase information over the atoms determines the directionality of the idler photon, i.e., a phase-matched direction, as a result of the constructive interference in the erasure process [17]. Therefore, the probability amplitude of the heralded photons is described as the

*hsmoon@pusan.ac.kr

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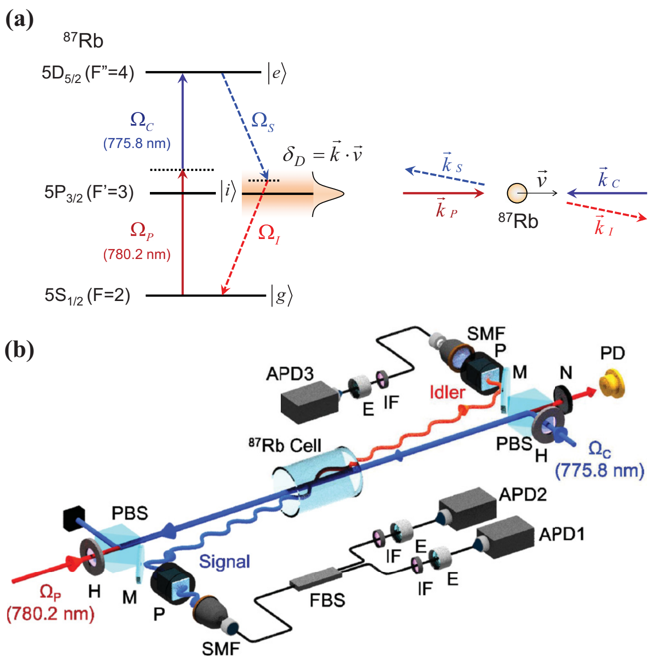

FIG. 1. (a) Cascade emission of signal and idler photons via spontaneous four-wave mixing in the Doppler-broadened four-level ladder-type atomic system. (b) Schematic of our experimental setup. The counterpropagating pump (Ω_P) and coupling (Ω_C) fields generate coherent two-photon excitation in all velocity groups, leading to photon-pair generation in the phase-matched direction.

coherent superposition of the photon pairs generated from the Doppler-distributed moving atoms.

The experimental setup for heralded-single-photon generation in the Doppler-broadened ladder-type atomic system is shown in Fig. 1(b). To achieve SPS in a Doppler-broadened atomic system, it is necessary to generate Doppler-free two-photon coherence for most of the moving atoms. In our study, the Ω_P and Ω_C laser fields were counterpropagated through a 12.5-mm-long vapor cell containing the ^{87}Rb isotope and spatially overlapped completely with the same $1/e^2$ beam diameters of 1.2 mm. The powers of the Ω_P and Ω_C fields were 1 and 10 mW, respectively, which were adjusted with the use of a half-wave plate and polarizing beam splitter (PBS). The temperature of the main cell was maintained at 52 °C, and the vapor cell was housed in a three-layer μ-metal chamber to shield against external magnetic fields. The signal and idler photons were generated in the phase-matched direction and collected in two single-mode fibers (SMFs).

To isolate the signal and idler photons from uncorrelated fluorescence, we used an interference filter with a 3-nm bandwidth and 95% transmittance as well as a solid fused-silica etalon filter with 950-MHz linewidth and 85% peak transmission. After passing through the interference filters and etalon filters, the photons were coupled into multimode fibers. The estimated total efficiency for obtaining spatially single-mode and noise-suppressed photon pairs was approximately 50%. The photons were detected by silicon avalanche photodetectors (APDs) [18] with quantum efficiency: ∼40%; detection-time jitter: ∼300 ps; dead time: ∼50 ns; measured dark count rates: 100 − 300 Hz.

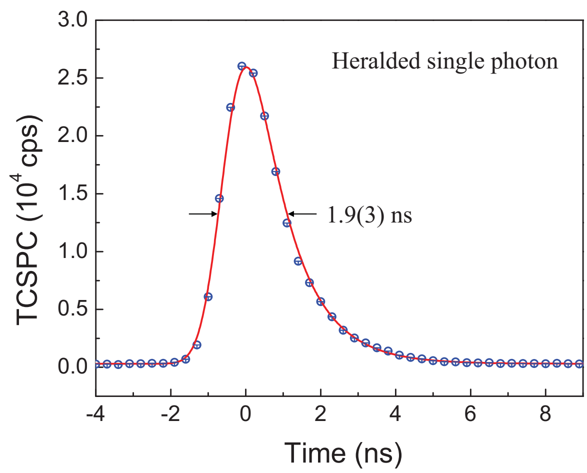

FIG. 2. Time-correlated single-photon counting (TCSPC) histogram of heralded single photons upon the conditional detection of a signal photon. Here, TCSPC histograms were measured with 4-ps time resolution for a total data-acquisition time of 30 s.

The characteristics of the photon pair were investigated with the use of the conditional Hanbury Brown–Twiss (HBT) experiment [16,19]. The apparatus included a fiber beam splitter and a time-correlated single-photon counting module [(TCSPC) PicoHarp 300] in the start-stop mode with a 4-ps time resolution. The normalized second-order correlation function $g_C^{(2)}(0)$ of a heralded single photon was measured to be 0.037(3) [16].

We investigated the properties of the heralded single photon in the Doppler-broadened atomic ensemble of the warm ^{87}Rb vapor cell. The temporal property of the heralded single photon was measured, as shown in Fig. 2. Figure 2 shows the temporal detection histogram of a heralded single photon, i.e., the cross-correlation function between the signal and idler photons. The spontaneous emission time of the heralded single photon was estimated to be 1.9(3) ns, which is significantly shorter than the excited-state lifetime of the ^{87}Rb atom. This shortened emission time of the heralded single photon is not understood as the spectral broadening due to the Doppler effect, because the spontaneous emission time of individual atoms in the inhomogeneous broadened media is never changed, even though its spectral width is broadened due to the Doppler effect. This shortened emission time results from the constructive interference between the superposed photonic amplitudes in different velocity groups, which is distinct from the dephasing of the spatial coherence induced by atomic motion. In our experiment, although the spontaneous emission time originates from the inhomogeneously broadened atoms, these atoms act as a single, large atom, radiating a single photon in a burst with a decay time of 1.9 ns, because of the collective effect of the Doppler-broadened atomic ensemble.

The red solid line in Fig. 2 indicates the fitted curve [20], where the height and offset are free parameters. The theoretical result should be considered as the collective effect of the Doppler-broadened atomic ensemble; the almost-velocity classes can coherently contribute to the generation of photon pairs. The experiment result is in good agreement with the theoretical curve. Therefore, we confirm that the spontaneous

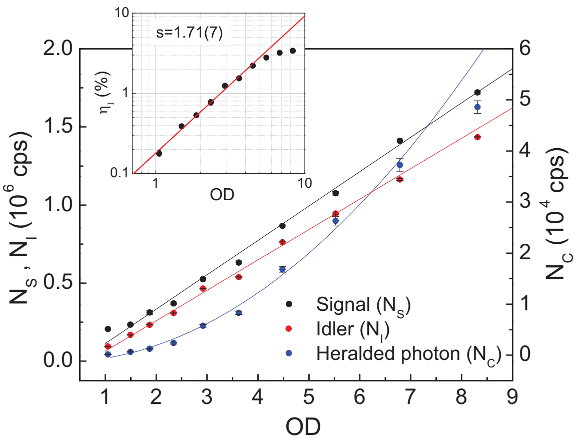

FIG. 3. The photon count rates for the signal (N_S) and idler (N_I) photons and the heralded-single-photon count rate (N_C) measured as functions of the optical depth (d_o). The black and red solid lines indicate linear fits, and the blue solid curve is a square fit. All fitting curves were based on measurements performed until the d_o was less than 7. Inset: Heralding efficiency as a function of d_o on a log-log scale. The red line in the inset graph indicates a linear fit with slope s = 1.71(7).

emission time of the heralded single photon is owing to the constructive interference between the two-photon amplitudes from different velocity groups.

The superradiance intensity is proportional to the square of the particle number, whereas the intensity of spontaneous radiation from individual atoms is linearly proportional to the number of atoms [1]. The evidence for SPS is the quadratic increment of the photon emission rate as a function of the atom number. The rate N_C that varies with the square of the number of atoms is related to the characteristic of the spontaneous four-wave mixing process. We investigated the relationship between the emitted-photon count rates and the atom number per unit volume by varying the vapor-cell temperature from 31 °C to 52 °C. Figure 3 shows the signal (N_S: black circles) and idler (N_I: red circles) photon count rates and the heralded-single-photon count rate (N_C: blue circles) as functions of the optical depth (d_o), wherein the signal- and idler-photon count rates are linearly proportional to the particle number [21]. As d_o increases from 1 to 7, N_S and N_I increase linearly. However, N_C is proportional to the square of the particle number; the increase in N_C is quadratic. The rate N_C that varies with the square of the number of atoms is related to the characteristic of the spontaneous four-wave mixing process. Here, we note that N_C represents the net coincidence counting rate minus random coincidences. When the detection probability (η_I) of a heralded single photon in the idler mode is plotted against d_o in a log-log graph (Fig. 3, inset), the accelerated emission of a heralded single photon can be expressed as $\eta_I \propto d_o^S$ with slope $s = 1.71(7)$. The quadratic tendency reduces at a high d_o of >7; this is primarily due to reabsorption of the idler photons in the atomic ensemble.

When the value of d_o increased more than 40, interestingly, we observed the appearance of an oscillation in the coincidence-event histogram within a time scale of several nanoseconds, as shown in Fig. 4(a). To interpret this observed

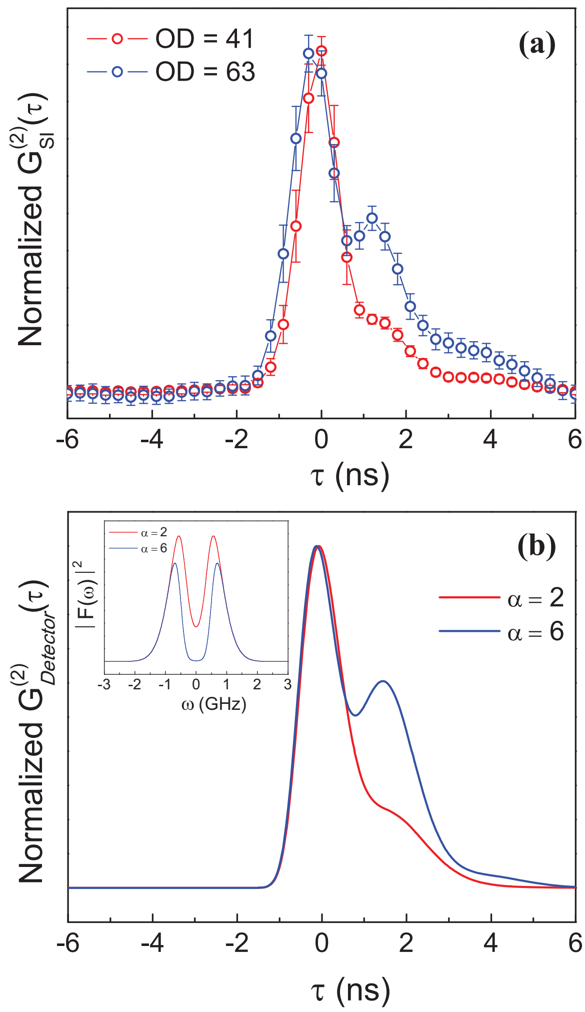

FIG. 4. (a) Normalized temporal histogram of coincidence events at $d_o = 41$and 63. (b) Plots of normalized second-order correlation functions on the detector $G^{(2)}_{\text{Detector}}(\tau)$, considering the corresponding filtering functions $F(\omega)$. Inset: Plots of $|F(\omega)|^2$ for absorption coefficient $\alpha = 2$ and 6.

oscillation, we assumed that the cause of oscillation is the beat between both two-photon amplitudes of the photon pairs generated from the two separated atomic velocity groups. It is noteworthy that the atomic ensemble is not only a photon emitter but also an effective photon absorber, particularly for idler photons, because the idler photons are resonant for the $5S_{1/2}$-$5P_{3/2}$ transition. After the idler photons are generated in the atomic ensemble, they can be significantly reabsorbed while passing through the highly dense atomic medium. As the absorption of idler photons becomes dominant at the $5S_{1/2}$-$5P_{3/2}$ transition resonance, the spectral feature of the idler photons coupled to the SMF can be distorted considerably from the Doppler profile of a warm ^{87}Rb vapor.

To theoretically analyze the experimental results shown in Fig. 4(a), we present a model in which the observed oscillation originates from the interference between wavelength-separated two-photon amplitudes via the reabsorption filtering effect. For a spontaneous four-wave mixing process in a Doppler-broadened four-level ladder-type atomic system, the biphoton wave packet is given by

$$\Psi_v(\tau) = A(v)e^{[-\Gamma/2+ik_I v]\tau}, \tag{2}$$

where Γ represents the decay rate of the $5S_{1/2}$-$5P_{3/2}$ transition, k_I the wave vector of an idler photon, v the atomic velocity, and τ the detection time difference between the signal and idler photons [20]. Here, $A(v)$ represents a coefficient depending on the atomic velocity, indicating the extent of the two-photon coherence generated in each velocity group.

The second-order cross-correlation function for the paired photons generated from a Doppler-broadened atomic ensemble is defined as

$$G^{(2)}_{SI}(\tau) = \left| \int \Psi_v(\tau) f(v) dv \right|^2, \tag{3}$$

where $f(v)$ denotes a one-dimensional Maxwell-Boltzmann velocity distribution function. Because almost all the velocity groups in an ensemble can coherently contribute to photon-pair generation, $G^{(2)}_{SI}(\tau)$ is significantly enhanced via constructive interference between the two-photon amplitudes from the different velocity groups.

Since the idler photons with frequencies within the Doppler bandwidth are filtered after the generation process, we can express the filtering function as [22,23]

$$F(\omega) = T(\omega) \exp[-\alpha A(\omega)], \tag{4}$$

where α denotes the absorption coefficient of $F(\omega)$, $T(\omega)$ a Gaussian function applied to an etalon filter with a 940-MHz bandwidth, and $A(\omega)$ the Gaussian function of the Doppler-absorption profile with a 540-MHz bandwidth.

At high d_o, the remaining idler photons emerge from the atomic velocity groups at far red- and blue-detuned wavelengths, as shown in the spectrum in the inset of Fig. 4(b). As for the uncorrelated photon emission and reabsorption, i.e., a typical spontaneous emission, since both processes are symmetric, it would hardly appear that only the center is absorbing whereas the periphery of the Doppler-broadened spectrum is emitting. The generation of correlated photons strongly depends on the two-photon coherence of the atomic ensemble which can be formed over the entire Doppler-broadened spectrum in our experimental configuration. On the other hand, the reabsorption process is not related to two-photon coherence and mostly occurs in the vicinity of resonance. A plot of $|F(\omega)|^2$ is shown in the inset of Fig. 4(b) for $\alpha = 2$ and 6. Here, we note that α is not the d_o under these experimental conditions; rather, it is a calculation parameter. Because the filtering effect on the electric-field operator in the time domain can be expressed by [24]

$$\hat{E}_{\text{Detector}}(t) = \int dt \tilde{F}(t-t')\hat{E}(t'), \tag{5}$$

where $\hat{E}(t)$ denotes the electric-field operator at time t and $\tilde{F}(t)$ the Fourier transform of $F(\omega)$, the final second-order cross-correlation function of $G^{(2)}_{\text{Detector}}(\tau)$ can be obtained through the simple convolution of $G^{(2)}_{SI}(\tau)$ and $|\tilde{F}(t)|^2$ as

$$G^{(2)}_{\text{Detector}}(t) = \int d\tau |\tilde{F}(t-\tau)|^2 G^{(2)}_{SI}(\tau). \tag{6}$$

The calculation results are plotted in Fig. 4(b); they exhibit suitable qualitative agreement with the experimental results. In order to qualitatively illuminate the quantum interference between elements of different frequencies, we considered the only reabsorption rate as we increase the value of d_o. However, as the value of d_o increases, the heralded-single-photon counting rate is proportional to the square of the particle number and the idler photon count rates increase linearly. Thus the inset of Fig. 4(b) should not be considered as a simple reabsorption spectrum of idler photon, but the combination with photon generation rate. In addition, d_o has been measured by fitting a linear absorption spectrum of a weak laser light which passes through the whole length of the vapor cell. On the other hand, most of the idler photons may not propagate through the whole vapor cell when we assume that the photon generation probability is equally distributed over the entire overlap region of pump lights in the vapor cell.

Therefore, we can conclude that the oscillating behavior is due to the interference between elements of different frequencies, i.e., the right- and left-hand sides of the strong absorption dip. Furthermore, as a number of different atomic velocity classes are coherently excited, the interference between the emissions of phase-matched optical dipoles belonging to different velocity classes leads to the heralded SPS beating in the temporal histogram. The observation of the heralded SPS beating is important evidence of the coherent superposition of the two-photon amplitudes from different velocity classes in the Doppler-broadened atomic ensemble.

In conclusion, we observed the heralded-single-photon superradiant beating in a Doppler-broadened ladder-type ^{87}Rb atomic system. The SPS effects in the Doppler-broadened atomic medium are closely related to the Doppler-free two-photon coherence via the interaction of the atomic ensemble with two coherent electromagnetic fields. The observation of single-photon superradiant beating under high-d_o conditions in the Doppler-broadened atomic medium indicates coherent superposition and interference between two-photon amplitudes belonging to different velocity classes. The heralded SPS in a Doppler-broadened atomic ensemble acts as single-photon spontaneous radiation from a single, large atom, which is composed of identical atoms with a Doppler-broadened linewidth. The importance of our work lies not only in the extended interpretation of superradiance in inhomogeneous broadened media, but also in the experimental confirmation of the collective behavior of moving atoms in the emission of heralded single photons. We believe that the SPS in the atomic vapor cell will facilitate the development of a method for producing a robust and bright photon-pair source with a relatively narrow bandwidth.

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