

## Second-order correlations in directed emissions in sodium atoms

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We report second-order intensity correlations  $g^{(2)}(\tau)$  of directional midinfrared emissions from sodium vapor under bichromatic continuous-wave excitation. Directional emission at  $2.34\ \mu\text{m}$  in the forward direction and at  $2.21\ \mu\text{m}$  in both forward and backward directions exhibits  $g^{(2)}(0)$  values that exceed unity yet remain well below the thermal limit of 2, indicating subthermal photon bunching characteristics of a multimode collectively driven process rather than purely independent amplified spontaneous emission. Cross-correlation measurements reveal that the counterpropagating  $2.21\text{-}\mu\text{m}$  beams are positively correlated, demonstrating dynamic coupling between the two emission channels. Persistent oscillations in  $g^{(2)}(\tau)$ , observed across a broad range of pump powers, shift systematically to higher frequencies with increasing excitation power, consistent with ac Stark shifts of hyperfine sublevels. Together, these observations establish that the emission arises from a phase-matched, continuous-wave collective process combining features of mirrorless lasing and amplified spontaneous emission, with long-range dipole coherence and velocity-selective coupling governing the photon statistics and the forward-backward emission symmetry. The experiment uses a 10-cm buffer-gas-free sodium vapor cell under bichromatic excitation at 589.2 and 569.0 nm; the emitted midinfrared light is detected in a Hanbury Brown–Twiss configuration. The demonstrated backward emission is of particular interest for applications in laser guide star generation and mesospheric remote sensing, where understanding the statistical properties of the emitted light is essential for optimizing sodium-based light sources.

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### I. INTRODUCTION

Coherent and nonlinear optical processes in alkali metal vapors provide a versatile platform for studying light-matter interactions in resonant media [1–3]. Among these processes, mirrorless lasing (ML) occupies a distinct position: directional coherent emission emerges without an external cavity, with optical feedback provided by amplified spontaneous emission (ASE) or distributed gain within the medium it-

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self [4–11]. In practice, ML is characterized by directed emission with a distinct pump threshold and spectral narrowing, although in the literature the term is sometimes used broadly to encompass stimulated emission processes (including ASE) operating in a regime of relatively short temporal coherence compared to conventional cavity-based lasers [12]. Owing to its alignment-free geometry and intrinsic directionality, ML is attractive for remote sensing, metrology, and laser guide star applications [13–16].

Mirrorless lasing has been demonstrated in several alkali metal vapors under both pulsed and continuous-wave excitation [4,17–19]. In rubidium and sodium vapors, directional midinfrared emission has been observed under resonant stepwise or two-photon excitation, including backward-propagating continuous-wave radiation at 2.21  $\mu\text{m}$  in sodium with a sub-10-mW threshold [20,21]. In addition, polychromatic emission involving infrared and blue components has been reported in cesium [22]. Beyond alkali metal vapors, directional amplified emission without an external cavity has been investigated in a wider class of systems. In particular, signal amplification in population-inverted media was established in early 1970s [23], which showed that threshold behavior and spectral narrowing are generic features of ASE-driven gain rather than signatures unique to cavity-based lasing. Continuous-wave mirrorless lasing has been demonstrated in mercury vapor under resonant two-photon pumping [24], extending the phenomenon beyond alkali metal systems. These precedents motivate careful discrimination between ASE-driven gain and more collective emission mechanisms—a central goal of the present work. While some of these emissions are consistent with ASE-driven gain, others involve parametric processes such as four-wave mixing (FWM), where no population inversion is present. Recent theoretical studies further suggest that collective dipole-dipole interactions and radiation-mediated correlations can support directional, spectrally narrowed emission in free space, even without a conventional cavity [25–27].

A direct way to characterize the statistical and coherence properties of such radiation is through the second-order intensity correlation function [28]

$$g_{1,2}^{(2)}(\tau) = \frac{\langle I_1(t) I_2(t + \tau) \rangle}{\langle I_1 \rangle \langle I_2 \rangle}, \quad (1)$$

where  $I_1$  and  $I_2$  are the intensities recorded in two detection channels, e.g., in a Hanbury Brown–Twiss configuration [29,30]. In the particular case of autocorrelation, a single source is considered and the correlation is evaluated between intensities at times  $t$  and  $t + \tau$ . The value  $g^{(2)}(0)$  distinguishes coherent ( $g^{(2)}(0) = 1$ ), thermal ( $g^{(2)}(0) = 2$ ), and antibunched ( $g^{(2)}(0) < 1$ ) light, and the functional form of  $g^{(2)}(\tau)$  provides access to coherence time and intensity dynamics. In alkali metal vapors, intensity correlations have been investigated mainly for forward-generated light and FWM signals [31], and oscillatory features in correlation functions have been associated with spiking or coherent population dynamics [32–36].

Despite extensive studies of directional emission and ML in alkali metal vapors, quantitative measurements of the second-order coherence for simultaneously generated forward- and backward-propagating midinfrared radiation

remain scarce. In particular, it is unclear whether the counterpropagating beams behave as independent ASE outputs or exhibit correlated dynamics that may be indicative of collective coupling within the gain medium.

In this work, we investigate the temporal intensity correlations of forward- and backward-propagating midinfrared emission at 2.21 and 2.34  $\mu\text{m}$  in sodium vapor under bichromatic continuous-wave excitation. By measuring both auto- and cross-correlation functions  $g^{(2)}(\tau)$ , we identify a persistent oscillatory structure and nontrivial correlations between the counterpropagating beams. These results demonstrate that the emission cannot be described solely as independent ASE, but instead involves collective coupling between longitudinal modes of the medium. Our measurements provide quantitative access to coherence time, threshold behavior, and intensity noise, and clarify the physical mechanism underlying directional midinfrared generation in sodium vapor, with direct relevance to bichromatic laser guide star schemes and remote sensing applications.

## II. EXPERIMENTAL SETUP AND PROCEDURES

The objective of the present experiment is to determine whether directional midinfrared emission from sodium vapor under continuous bichromatic excitation can be adequately described as independent amplified spontaneous emission, or whether it exhibits signatures of collective light-matter dynamics. To this end, we simultaneously detect the forward- and backward-propagating emissions at 2.21  $\mu\text{m}$ , and forward-propagating emission at 2.34  $\mu\text{m}$ , and analyze their temporal intensity correlations through measurements of the second-order correlation function  $g^{(2)}(\tau)$ . By comparing auto- and cross-correlation functions over a range of pump powers, we directly probe the coherence time, statistical properties, and mutual coupling of the counterpropagating fields.

The experimental setup is largely based on the one presented in Ref. [37]. Its simplified version, as well as relevant atomic transitions, are presented in Fig. 1.

The experiment employs a 10-cm-long sodium vapor cell without buffer gas. The cell is mounted inside a magnetic shield composed of four layers of  $\mu$ -metal to suppress stray magnetic fields; a resistive heater consisting of a pair of twisted wires provides a controlled temperature gradient along the cell while ensuring reduced magnetic-field noise. The temperature of the vapor cell is kept at 170  $^\circ\text{C}$  throughout the experiment. The wedge-shaped angled windows of the cell help suppress optical feedback.

Two laser systems are used for excitation: a solid-state Toptica TA-SHG Pro laser operating at 589.2 nm resonant with the  $3S_{1/2} \rightarrow 3P_{3/2}(F' = 3, 2, 1)$  transition, and a Coherent 899-21 dye laser at 569.0 nm, driving the  $3P_{3/2} \rightarrow 4D_{5/2}$  transition. Efficient ground-state repumping, which lowers the ASE threshold and enhances excitation efficiency [38], is achieved by generating spectral sidebands at 1.713 GHz via frequency modulation of the 589.2-nm laser using an electro-optic modulator with a modulation depth of  $-12$  dBm. The outputs of the two lasers are combined on a nonpolarizing beam splitter to form a bichromatic excitation beam, which is directed into the sodium cell with a minimum spot diameter of approximately 1 mm. The polarization and intensity of both

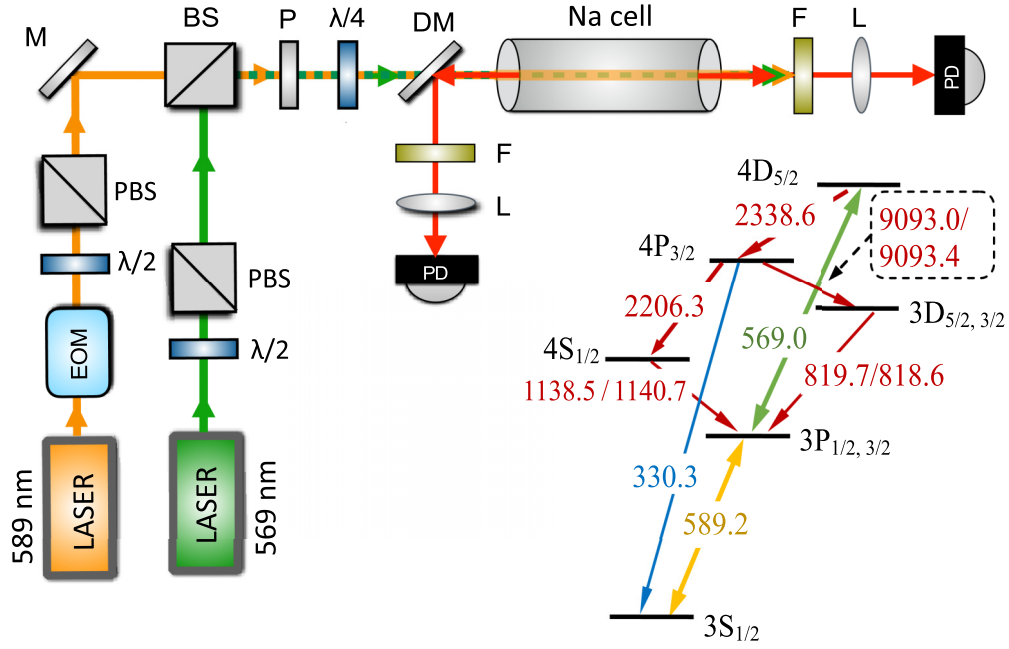


FIG. 1. Schematic top view of the experimental setup on the left. EOM, electro-optic modulator driven at 1.713 GHz; PBS, polarizing beam splitter; BS, nonpolarizing beam splitter; M, mirror; P, polarizer; DM, dichroic mirror; F, infrared filter at 2.21 or at 2.34  $\mu\text{m}$ ; L, lens; PD, photodetector working in the range 2–9  $\mu\text{m}$ . Relevant sodium energy levels and optical transitions are shown on the right. The atomic ensemble is driven by two continuous-wave laser fields at 589.2 and 569.0 nm, resonant with the  $3S_{1/2} \rightarrow 3P_{3/2}(F' = 3, 2, 1)$  and  $3P_{3/2} \rightarrow 4D_{5/2}$  transitions, respectively, thereby populating the  $4D_{5/2}$  excited state. The driving laser fields are circularly polarized at the entrance of the cell in the same way. Spontaneous radiative decay channels from the excited states are indicated by red arrows for infrared transitions and by a blue arrow for the ultraviolet emission at 330 nm. In the present work, we investigate the directional midinfrared emissions at 2.21 and 2.34  $\mu\text{m}$ , which are detected simultaneously in the forward and backward directions.

the 589.2- and 569.0-nm components are controlled using wave plates and polarizers. We ensure both driving laser fields have circular polarization.

Midinfrared emission directed along the laser propagation axis is spectrally selected using filters centered at 2.21 or 2.34  $\mu\text{m}$  and detected with a VIGO LabM-I-10.6 photodetector (operating in the 2–9- $\mu\text{m}$  range). The signal is recorded using a data acquisition (DAQ) system with a sampling rate of 500 MS/s. We also observe backward emission at 2.21 and 2.34  $\mu\text{m}$ . However, the intensity of the 2.34- $\mu\text{m}$  backward emission is too weak to be reliably detected and analyzed at the same experimental conditions, and therefore it is not further considered in this work. Forward and backward emissions at 2.21  $\mu\text{m}$  are simultaneously measured using two PDs, with each radiation recorded on a two-channel DAQ card. For the laser light measurements, we employ a Thorlabs PDA10A photodetector, as the VIGO detector is not sensitive to the laser wavelengths used.

With 589.2-nm light at 230 mW and 569.0-nm light at 45 mW, we observe 80  $\mu\text{W}$  of the 2.21- $\mu\text{m}$  collimated light in the forward direction and 45  $\mu\text{W}$  in the backward direction. Under the same conditions, forward-emitted light at 2.34  $\mu\text{m}$  has the power of 12  $\mu\text{W}$ . Partial time traces of raw normalized intensities of forward- and backward-directed emissions at 2.21  $\mu\text{m}$  are shown in Fig. 2(a) in solid and dashed lines, respectively, revealing periodic spike structure and pronounced oscillations (see also Appendix A).

Using the measured laser powers and estimated beam diameters, we find that both excitation lasers operate well above saturation (see Appendix B). Order-of-magnitude estimates based on known dipole matrix elements yield Rabi frequencies of around 1 GHz for the 589.2-nm transition and of order 200 MHz for the 569.0-nm transition, exceeding the natural linewidths by more than an order of magnitude. These estimates confirm that the observed dynamics correspond to a regime of strong optical driving combined with weak emitted fields.

### III. RESULTS AND DISCUSSIONS

To verify the approach for extracting the second-order correlation functions, we first measure the intensity of cw lasers at 569.0 and 589.2 nm. All measurements were carried out over a total acquisition time of 500 ms. The recorded intensity traces were partitioned into 1000 equal segments, and the second-order correlation function  $g^{(2)}(\tau)$  was evaluated for each segment using Eq. (1) and then averaged. Calculated  $g^{(2)}(\tau)$  functions are shown in dash-dotted green and solid yellow lines, respectively, in Figs. 2(b), 2(c), and 2(d). The deviation of  $g^{(2)}(0)$  from the expected value of 1 is less than  $10^{-4}$  for both lasers.

With the same technique (but using the VIGO LabM-I-10.6 detector), we recorded the emission at 2.21  $\mu\text{m}$  in both forward- and backward- directions simultaneously, as well as the forward-directed emission at 2.34  $\mu\text{m}$ . Fig-

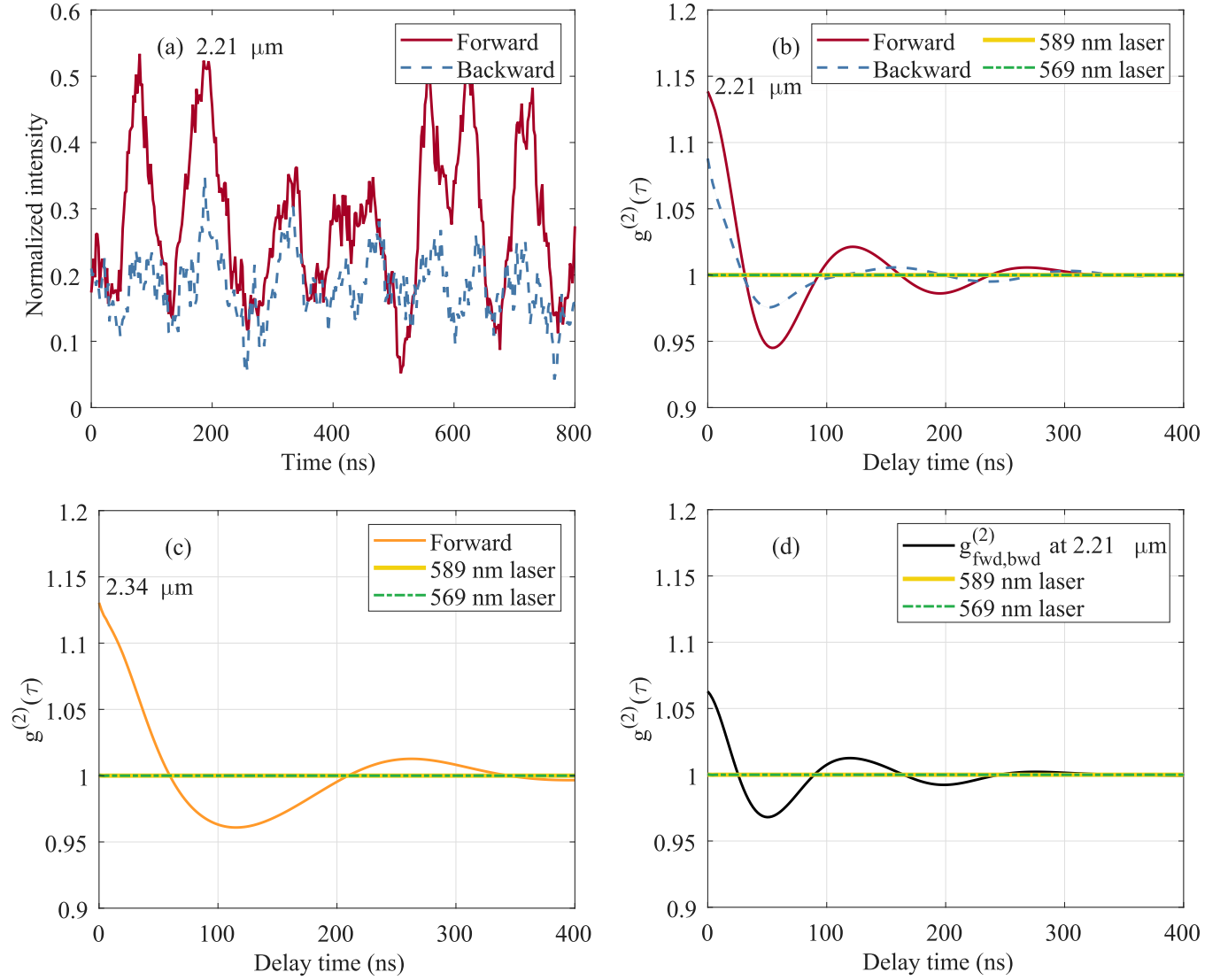


FIG. 2. (a) Partial time traces of raw normalized intensities of forward- and backward-directed emissions in solid and dashed lines, respectively, revealing periodic spike structure. (b) Autocorrelation functions vs delay time  $\tau$  for simultaneously detected forward (solid) and backward (dashed) emissions at 2.21  $\mu\text{m}$ . (c) Autocorrelation function vs delay time  $\tau$  for forward emission at 2.34  $\mu\text{m}$ . (d) Cross-correlation vs delay time  $\tau$  in solid line between forward- and backward-emitted lights at 2.21  $\mu\text{m}$ . The 569.0-nm laser power is 15 mW, and the 589.2-nm laser power is 230 mW. Autocorrelation functions vs delay time  $\tau$  for the 589.2-nm laser (solid horizontal line) and 569.0-nm laser (dash-dotted horizontal line).

ure 2(b) shows the results for the 2.21- $\mu\text{m}$  emission, with the forward-directed radiation represented by a solid line and the backward-directed radiation by a dashed line. We note that the  $g^{(2)}(\tau)$  functions are smoother than the raw intensity traces. This is because  $g^{(2)}(\tau)$  probes correlations of intensity fluctuations rather than their instantaneous amplitude. Consequently, rapidly varying fluctuations that are prominent in the instantaneous intensity traces are substantially suppressed in the correlation function. Additional averaging over multiple temporal segments further reduces statistical fluctuations. Figure 2(c) displays the forward-directed emission at 2.34  $\mu\text{m}$  as a solid line. The standard errors are smaller than the line thickness and are invisible in the figures. An example of  $g^{(2)}(\tau)$  calculation from a single segment is presented in Appendix A. All data in Fig. 2 correspond to an incident pump power of 15 mW at 569.0 nm and 230 mW at 589.2 nm. As

seen in the figure,  $g^{(2)}(0) > 1$ , it indicates photon bunching and demonstrates that the observed “mirrorless lasing” does not correspond to pure lasing, but rather contains at least an admixture of ASE. Using Eq. (3.8.7) from Ref. [28], the expected value of  $g^{(2)}(0)$  for a single-mode signal is anticipated to be approximately  $\approx 1.78$  for chaotic light, given a coherence time of  $\approx 20$  ns and an integration time of 8 ns. However, our measured value is significantly lower.

On the other hand, subthermal values  $1 < g^{(2)}(0) < 2$  can arise from the detection of multiple independent spatiotemporal modes and finite measurement bandwidth (integration time) [39]. For mutually incoherent chaotic modes, one expects  $g^{(2)}(0) = 1 + 1/M$ , where  $M$  is the number of effective modes [40]. The calculated cross-correlations between the 2.21- $\mu\text{m}$  forward- and backward-emitted radiations are shown in Fig. 2(d) with a solid curve. Again, the cross-correlation

between the forward- and backward-emitted 2.21- $\mu\text{m}$  light takes values between 1 and 2.

The observed positive cross-correlation between the forward- and backward-propagating channels is nontrivial. Independent ASE processes would be expected to produce statistically uncorrelated emission; competition between forward and backward emissions would produce anticorrelation. The measured positive correlation at  $\tau = 0$  therefore indicates that the two directions are dynamically coupled. We attribute this behavior to the interplay of several nonlinear mechanisms in the driven sodium vapor [41,42]. These may include pump-induced refractive-index modifications, redistribution of population among distinct atomic velocity groups [43], and the participation of the forward-propagating field in additional four-wave mixing loops. A quantitative description and analysis of these coupled processes is beyond the scope of the present work. Here we limit ourselves to an experimental characterization of the observed correlations.

In the dependence on delay time of autocorrelations and cross-correlations, oscillatory behavior is observed. A pronounced oscillation with a period of around 120 ns, which is equivalent to 8.3 MHz, is already apparent from the signal structure itself, which is translated to the time dependence of  $g^{(2)}(\tau)$  [see Fig. 2(b), solid line]. Additional, less obvious components are deduced using the Fourier transform of the signal and the corresponding  $g^{(2)}(\tau)$  function, which yielded consistent spectral features. Oscillation frequencies are presented in Fig. 3 as a function of 569.0-nm laser power, and 589.2-nm laser power is fixed at 230 mW.

Across the range of pump powers investigated, we observe persistent oscillations around 4–5, 8–11, and 12–16 MHz (see Fig. 3). The systematic upshift of these frequencies with increasing power is consistent with light-induced (ac Stark) shifts of the relevant energy splittings (see Appendix C). In addition, because the Rabi frequencies for both excitation transitions remain on the order of or smaller than the Doppler width of the optical transitions ( $\Delta\nu_D$  is a few GHz under our experimental conditions), the atom-light interaction is velocity selective. Excitation to the  $4D_{5/2}$  state therefore occurs predominantly for atoms whose velocity projection along the pump-beam propagation direction satisfies the two-photon resonance condition. This leads to a velocity-selective population inversion and gain on subsequent transitions in the cascade, in particular  $4D_{5/2} \rightarrow 4P_{3/2} \rightarrow 4S_{1/2}$ .

Another possible reason for oscillations is the dynamical buildup of emission, arising from the competition between time-coherent mirrorless lasing and ASE processes [32,33], although in contrast to the results reported in these papers, we do not observe damping of the oscillations far away from the emission threshold.

Figure 4 summarizes the dependence of several statistical properties of the forward- and backward-emitted radiation on the 569.0-nm pump power. The 589.2-nm laser power was held constant at 230 mW. The left column corresponds to emission at 2.21  $\mu\text{m}$  and the right column to emission at 2.34  $\mu\text{m}$ . In each column, the top panel displays the coherence time  $\tau_c$ , the middle panel shows the zero-delay intensity autocorrelation  $g^{(2)}(0)$ , and the bottom panel reports the normalized mean intensity of the detected directional emission. The intensities (bottom panels) are normalized to the maxi-

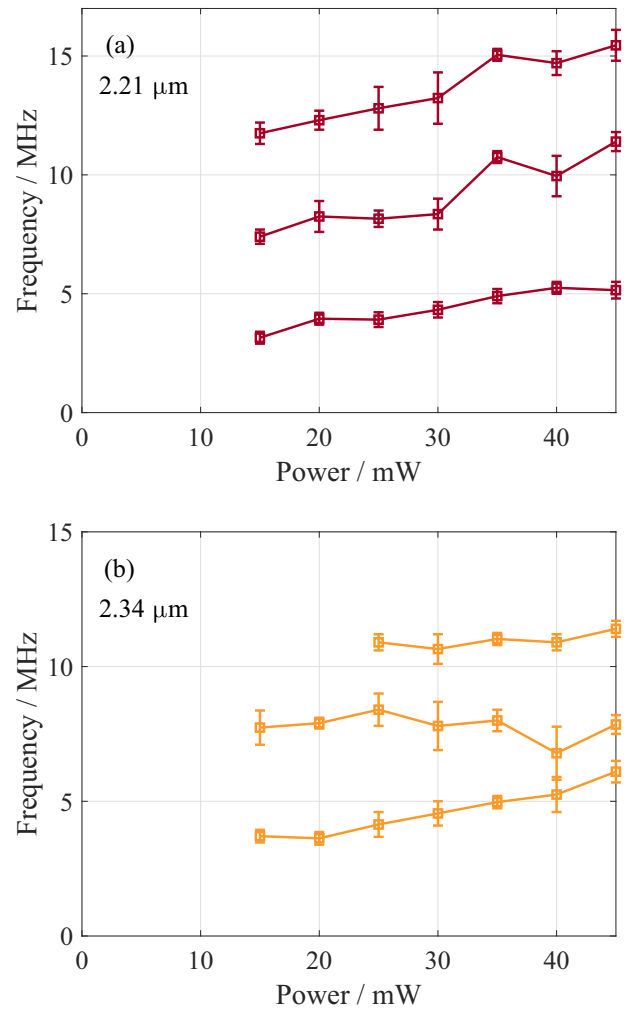


FIG. 3. Dependence of the oscillation frequencies extracted from the Fourier spectra of the intensity spike structure of the emitted radiation on the 569.0-nm laser power for forward-emitted 2.21  $\mu\text{m}$  (a) and 2.34  $\mu\text{m}$  (b) fields. Three persistent spectral components are identified (shown as separate symbol or line series, at around 4–5, 8–11, and 12–16 MHz), each shifting to higher frequency with increasing pump power, consistent with ac Stark shifts of the relevant hyperfine sublevels. Error bars reflect the uncertainty in identifying peak positions in the Fourier spectrum. The 589.2-nm laser power is fixed at 230 mW.

imum forward-emitted intensity measured at each wavelength. Specifically, the 2.21- $\mu\text{m}$  data are normalized to the maximum forward-directed intensity at that wavelength, and the 2.34- $\mu\text{m}$  data are normalized analogously. In both cases, the maximum occurs at a 569.0-nm laser power of 45 mW.

For the 2.21- $\mu\text{m}$  radiation, we plot forward-directed and backward-directed emissions for direct comparison (solid and dotted traces, respectively); the 2.34- $\mu\text{m}$  emission is shown in the right column. All quantitative fits shown in Fig. 4 include statistical uncertainties. The y-axis uncertainties lie within the symbol size and are therefore not visible, whereas the uncertainty in the laser power is estimated as  $\pm 0.7$  mW, and this is reflected in the horizontal error bars of the plots.

All measured beams exhibit qualitatively similar behavior as a function of the 569.0-nm pump power: no directional

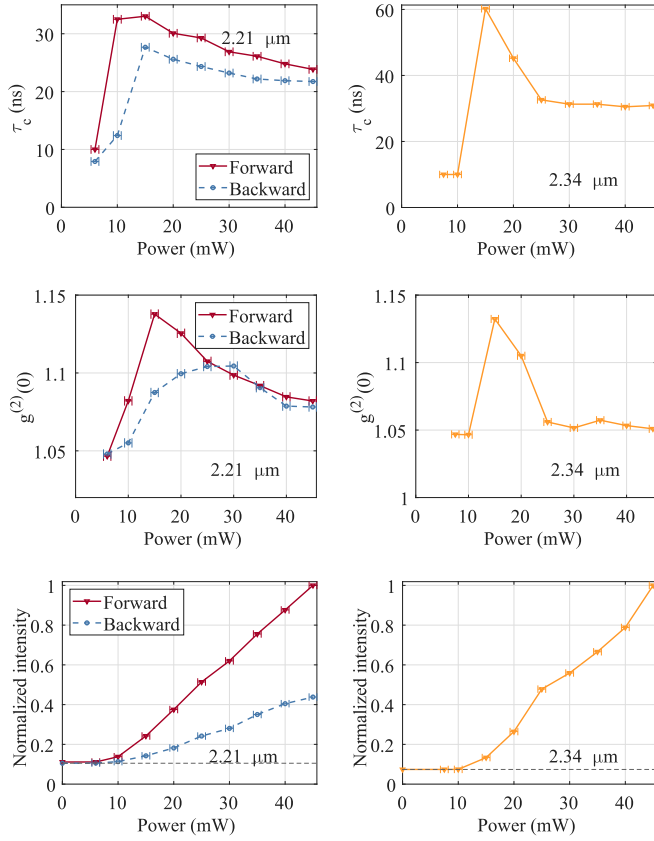


FIG. 4. Dependence on the 569.0-nm laser power of the coherence time (top panels), the zero-delay second-order correlation function  $g^{(2)}(0)$  (middle panels), and the normalized mean intensity (bottom panels) of the emitted midinfrared radiation (normalization is to the maximum of forward-emitted intensity measured at each wavelength). The left column corresponds to the 2.21- $\mu\text{m}$  emission and the right column to the 2.34- $\mu\text{m}$  emission. Solid lines denote forward emission, and dashed lines denote backward emission. The 589.2-nm excitation power is fixed at 230 mW. Horizontal error bars indicate the uncertainty in the 569.0-nm laser power ( $\pm 0.7$  mW) due to power fluctuations.

emission is detected below a pump threshold near  $P_{569} \approx 7$  mW, while above the threshold, the signals grow and their statistical properties evolve. We note that several different nonlinear optical mechanisms can display thresholdlike onsets (e.g., ASE, FWM, and ML).

The coherence time  $\tau_c$  was extracted by fitting the measured correlation function  $g^{(2)}(\tau)$  with a Gaussian profile and taking the  $1/e$  decay width as  $\tau_c$  [28]. The coherence time reaches a maximum at  $P_{569} \approx 15$  mW for all recorded beams, and for larger powers, it approaches approximately 20 ns for the 2.21- $\mu\text{m}$  emission and  $\approx 30$  ns for the 2.34- $\mu\text{m}$  emission. The value of  $g^{(2)}(0)$  likewise evolves with 569.0-nm pump power and asymptotes to a nearly constant value: for the 2.34- $\mu\text{m}$  emission, this occurs already at  $P_{569} \approx 25$  mW, whereas for the 2.21- $\mu\text{m}$  emission, the asymptotic behavior is reached at higher pump powers,  $\approx 40$  mW. For the 2.21- $\mu\text{m}$  emission,  $g^{(2)}(0)$  rises from about 1.04 at threshold to a peak value of 1.14 before settling to 1.08 at higher powers, while for the 2.34- $\mu\text{m}$  emission, it follows a similar trend, peaking

at 1.13 and stabilizing near 1.05. Such small deviations above unity are consistent with nearly coherent (laserlike) emission, but with a weak excess of super-Poissonian fluctuations that likely arise from residual amplified spontaneous emission or parametric contributions.

The tendency of both  $\tau_c$  and  $g^{(2)}(0)$  to level off at high 569.0-nm pump power is consistent with the system reaching a steady-state regime in which the dominant gain and loss processes balance; this behavior is expected for both the chaotic source [ $g^{(2)}(0) = 2$ ] and the laser [ $g^{(2)}(0) = 1$ ].

The mean detected intensity of the directional emission exhibits a clear threshold near  $P_{569} \approx 10$  mW (see Fig. 4, bottom row). A pump threshold is a generic feature of gain-driven directional emission, including ASE, FWM, and ML, and does not by itself discriminate between these mechanisms. The onset observed near  $P_{569} \approx 7$ –10 mW therefore characterizes the beginning of gain-dominated directional emission, while the nature of that emission is instead revealed by the photon statistics and cross-correlation measurements presented here. Below this 7–10-mW value, the signal remains constant at the detector offset level (indicated by the thin black dashed line). Above the threshold, the intensity increases linearly with pump power, indicating the onset of gain-dominated directional emission. The two midinfrared channels at 2.21  $\mu\text{m}$  (forward and backward) and 2.34  $\mu\text{m}$  (forward) exhibit similar statistical behaviors.

#### IV. CONCLUSION

In summary, we have investigated the statistical properties of sodium emission under bichromatic excitation by analyzing the second-order intensity correlation function  $g^{(2)}(\tau)$ . The directional emissions at 2.21 and 2.34  $\mu\text{m}$  exhibit  $g^{(2)}(0)$  values between 1 and 2, together with strong forward-backward correlations and persistent oscillations, pointing to a phase-matched, steady-state collective emission regime. These observations are consistent with cw directional emission [44], in which synchronized dipoles radiate coherently into both counterpropagating modes. Such behavior naturally accommodates the measured subthermal statistics and the presence of multiple effective modes, as partial mode averaging reduces the observed bunching amplitude without destroying collective coherence.

Throughout the explored range of pump powers,  $g^{(2)}(\tau)$  of the emissions exhibited persistent oscillations at characteristic frequencies near 4–5, 8–11, and 12–16 MHz, with a systematic upshift of these components at higher excitation power. It is consistent with ac Stark shifts of the underlying hyperfine sublevels participating in the collective polarization.

This behavior can be placed in a broader context: in photon Bose-Einstein condensation the mode populations follow a Bose-Einstein distribution, and the growth of the macroscopic ground-state occupation is reflected in the second-order correlation function  $g^{(2)}(\tau)$ , which tracks the crossover from thermal to coherent behavior. Near threshold the condensate may still exhibit bunching with  $g^{(2)}(0) > 1$  due to reservoir-induced number fluctuations, while in a well-developed condensate the ideal expectation is  $g^{(2)}(0) \approx 1$ , signifying second-order coherence [45]. Although directional scattering and photon condensation arise from distinct physical

mechanisms—nonequilibrium amplification versus equilibrium condensation—both phenomena can be characterized by their photon correlations. In this sense,  $g^{(2)}(\tau)$  offers a unifying diagnostic for identifying thermal, amplified, and condensed regimes of light, even though the statistical origins of coherence differ.

In addition, the backward-emitted light offers promising opportunities for laser guide star generation and mesospheric remote sensing, and our  $g^{(2)}$  analysis provides valuable insight into the coherence and mode structure of this emission, paving the way for the optimization of sodium-based light sources for atmospheric and astronomical applications. Looking ahead, extending these studies to more complex excitation geometries and structured light fields could provide further insight into the interplay between coherence, polarization, and spatial mode structure. In particular, measuring quantum correlations in polychromatic radiation generated by twisted light carrying orbital angular momentum represents a promising direction for future research.

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### DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

### APPENDIX A: TEMPORAL STRUCTURE AND AUTO- AND CROSS-CORRELATION FUNCTIONS

In order to illustrate the temporal dynamics of the detected midinfrared emission, we present the raw intensity traces of the 2.21- $\mu\text{m}$  radiation in both forward and backward directions under excitation with 230 mW at 589.2 nm and 45 mW at 569.0 nm. The time-dependent intensities are shown in Fig. 5(a), with the forward-directed radiation plotted by solid and the backward-directed radiation by dashed line. The radiations clearly exhibit spikelike structures in both the forward and backward directions, which appear largely in phase, indicating a strong correlation between the two emissions. The mean of the signals of the forward and backward emissions is in the order of  $\approx 10^3$ , and variance is in the order of  $\approx 10^5$ .

The corresponding second-order correlation functions, calculated directly from these data, are presented in Fig. 5(b). Here, the dash-dotted line corresponds to the forward emis-

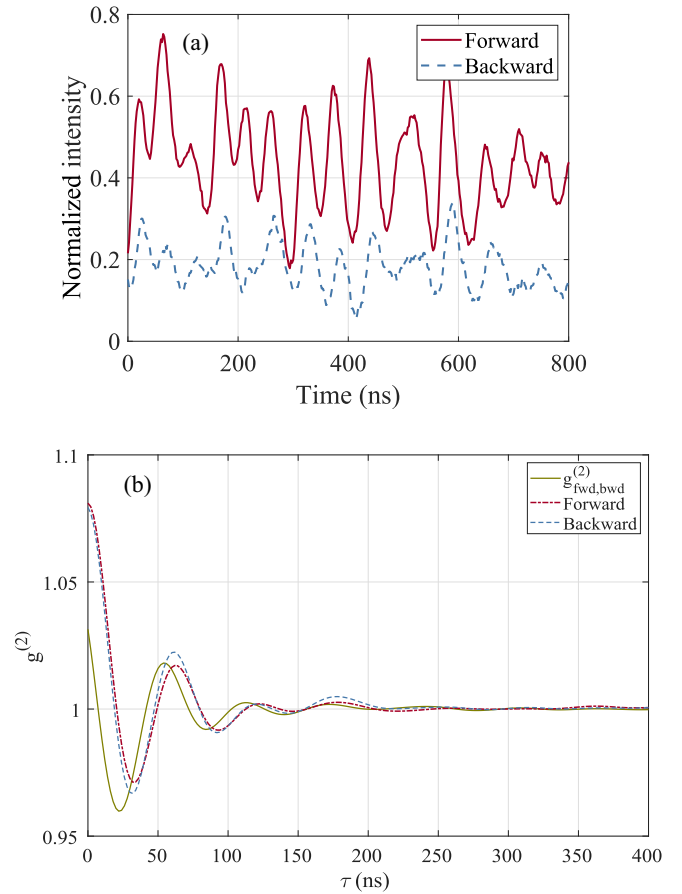


FIG. 5. (a) Time dependence of the detected intensities at 2.21  $\mu\text{m}$  in the forward (solid) and backward (dashed) directions, recorded with laser powers of 230 mW at 589.2 nm and 45 mW at 569.0 nm. (b) Corresponding second-order autocorrelation functions  $g^{(2)}(\tau)$  calculated from the same data: forward emission (dash-dotted line), backward emission (dashed line), and forward-backward cross-correlation (solid).

sion, the dashed line to the backward emission, and the solid curve to the cross-correlation between the two. These results provide a representative example of the raw data underlying the correlation analysis and demonstrate the presence of correlated fluctuations between forward- and backward-propagating emissions at 2.21  $\mu\text{m}$ .

### APPENDIX B: ESTIMATES OF INTENSITIES, AND RABI FREQUENCIES

#### 1. Intensity estimates

Intensities were estimated from the measured beam powers (230 and 45 mW) and diameters of approximately 1 mm for the excitation lasers at 589.2 and 569.0 nm. We estimate diameters of the generated midinfrared fields to be around 1 mm; however, these diameters were not directly measured and therefore introduce an uncertainty.

Using these values, we estimate peak intensities of  $I_{589} \approx 30 \text{ W/cm}^2$ ,  $I_{569} \approx 6 \text{ W/cm}^2$ . Both intensities exceed the corresponding saturation intensities by more than an order of

TABLE I. Reduced dipole matrix elements in atomic units and wavelengths for relevant sodium transitions used in the estimation of Rabi frequencies.

| Transition                          | Matrix element (a.u.) | Wavelength (nm) |
|-------------------------------------|-----------------------|-----------------|
| $3P_{3/2} \leftrightarrow 3S_{1/2}$ | 4.9838(34)            | 589.1583        |
| $3P_{1/2} \leftrightarrow 3S_{1/2}$ | 3.5246(23)            | 589.7558        |
| $4D_{5/2} \leftrightarrow 3P_{3/2}$ | 2.5705(86)            | 568.9783        |
| $4D_{5/2} \leftrightarrow 4P_{3/2}$ | 16.2356(81)           | 2338.55         |
| $4P_{3/2} \leftrightarrow 4S_{1/2}$ | 11.8360(59)           | 2206.25         |
| $4S_{1/2} \leftrightarrow 3P_{1/2}$ | 3.5762(18)            | 1138.46         |
| $4S_{1/2} \leftrightarrow 3P_{3/2}$ | 5.0670(25)            | 1140.69         |

magnitude, placing the system well within the strong-driving regime.

For the generated midinfrared radiation at 2.21 and 2.34  $\mu\text{m}$ , with measured output powers of order 80 and 120  $\mu\text{W}$  and an assumed diameter of 1 mm, the estimated intensities are  $I_{\text{IR}} \approx 10\text{--}15 \text{ mW/cm}^2$ .

## 2. Dipole matrix elements of relevant transitions

Table I summarizes the reduced dipole matrix elements used for the estimates below, taken from Ref. [46].

## 3. Estimated Rabi frequencies

Using the estimated intensities and the dipole matrix elements listed in Table I, we obtain order-of-magnitude estimates for the on-resonance Rabi frequencies. For the  $3S_{1/2} \rightarrow 3P_{3/2}$  transition at 589.2 nm, we estimate  $\Omega_{589} \simeq 2\pi \times 1 \text{ GHz}$ , while for the  $3P_{3/2} \rightarrow 4D_{5/2}$  transition at 569.0 nm we obtain  $\Omega_{569} \simeq 2\pi \times 200 \text{ MHz}$ . For the generated midinfrared fields at 2.21 and 2.34  $\mu\text{m}$ , the corresponding Rabi frequencies are estimated to be  $\Omega_{\text{IR}} \lesssim 2\pi \times 40\text{--}70 \text{ MHz}$ .

TABLE II. Estimated hyperfine splittings  $\Delta F$  for the  $4D_{3/2,5/2}$ ,  $4P_{1/2,3/2}$ , and  $4S_{1/2}$  states of sodium. The second-to-last row indicates transitions whose hyperfine separations correspond most closely to the oscillation frequencies observed in  $g^{(2)}(\tau)$ .

| $\Delta F$ | 1-0  | 2-1   | 3-2   | 4-3  |
|------------|------|-------|-------|------|
| $4D_{3/2}$ | 0.23 | 0.46  | 0.69  |      |
| $4D_{5/2}$ |      | 0.056 | 0.084 | 0.11 |
| $4P_{1/2}$ |      | 54    |       |      |
| $4P_{3/2}$ | 5.1  | 11.2  | 19.3  |      |
| $4S_{1/2}$ |      | 404   |       |      |

We emphasize that these estimates are approximate and rely on assumed beam diameters and uniform intensity profiles. Nevertheless, they are sufficient to identify the relevant dynamical regime and establish that the excitation lasers strongly saturate the driven transitions.

## APPENDIX C: HYPERFINE SPLITTINGS OF RELEVANT STATES

As discussed in the main text, the temporal structure of the registered emission signals reveals several characteristic frequency components. Across the investigated 569.0-nm laser power range, we observe persistent oscillations in  $g^{(2)}(\tau)$  around 4–5, 8–11, and 12–16 MHz. For comparison, the hyperfine splittings of the  $4D_{3/2,5/2}$ ,  $4P_{1/2,3/2}$ , and  $4S_{1/2}$  states were estimated using data from Refs. [47–49], and the corresponding energy separations  $\Delta F$  are summarized in Table II. The second-to-last row indicates the transitions whose hyperfine differences most closely match the observed oscillation frequencies, suggesting that these arise from beatings between the respective sublevels. Discrepancies can be attributed to ac Stark shifts, finite accuracy of hyperfine constants, and experimental factors such as detection bandwidth, residual magnetic fields, and multimode coupling.

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