Nonclassical Light from Large Ensembles of Trapped Ions

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(Received 19 May 2017; revised manuscript received 29 November 2017; published 20 June 2018)

The vast majority of physical objects we are dealing with are almost exclusively made of atoms. Because of their discrete level structure, single atoms have proved to be emitters of light, which is incompatible with the classical description of electromagnetic waves. We demonstrate this incompatibility for atomic fluorescence when scaling up the size of the source ensemble, which consists of trapped atomic ions, by several orders of magnitude. The presented measurements of nonclassical statistics on light unconditionally emitted from ensembles containing up to more than a thousand ions promise further scalability to much larger emitter numbers. The methodology can be applied to a broad range of experimental platforms focusing on the bare nonclassical character of single isolated emitters.

DOI: 10.1103/PhysRevLett.120.253602

In 1905, Albert Einstein discovered that light can be understood as a stream of interfering photons [1]. At that time, it was a contradiction to Maxwell's theory of light waves [2]. Quantum optics, however, merges our understanding of both wave and particle aspects together [3]. It seems to be commonly considered that light produced by a large number of emitters is expected to be a mixture of classical waves [4]. This has been certified by a number of experiments analyzing various light sources from sunlight to laser radiation, even with arbitrarily small intensity [5]. On the other hand, light from a single emitter with a transition between two discrete energy levels of an atom or solid-state object is always nonclassical. Detection of an individual photon emitted from a single emitter cannot be described by a mixture of classical waves, and therefore, many results of single-photon experiments contradict classical coherence theory [6]. The most basic contradiction is the observation of the indivisibility of a single photon at a beam splitter, which does not happen for classical light waves [7,8]. However, the situation is already different for two photons as they can be split by linear optics. It is evident that ideal particle indivisibility typically manifested by measurement of perfect antibunching in the case of single-photon input is lost for a large number of photons even though some particlelike nonclassical features may remain. On the other hand, the state of a finite number of photons cannot be modeled as a mixture of classical waves, as those are always based on distributions of photons up to infinity. This particle type of nonclassicality can remain hidden since the limitation in the number of photons can be far away from what is detectable, as can be seen on classical sources consisting of enormous amounts of quantum emitters.

Observability of truly particle aspects of light therefore typically vanishes in everyday macroscopic reality, and we frequently observe only mixtures of classical waves. This is due to the inevitable enhancement of effects, which deteriorate the macroscopic photon samples but also affect their macroscopic sources and detectors. However, those effects are not fundamental and macroscopicity itself does not smudge the particle features. In order to succeed with the detection of particlelike nonclassical aspects of light for large-number quantum emitters, the employed detection setup and evaluation procedure must provide unambiguous identification and sufficient information about nonclassical light without any prior assumptions. In addition, the measured light source needs to be stable in the number of photon emitters, and the detection efficiency of unconditionally emitted light has to be sufficiently high to overcome the effects of background thermal noise. The detection apparatus and the employed criterion should be able to detect nonclassicality for a large number of photons in the presence of high loss, finite amplitude of added thermal noise and within feasible measurement times. These requirements have long forced the particlelike nonclassicality of radiation from large ensembles of emitters and its possible applications to remain in the domain of theoretical considerations. Apart from a few notable recent stimuli, which provide substantial evidence that nonclassicality is not necessarily bound to a few emitters [9,10], experimental observations have been concerned by the analysis and control of nonclassical features on a very small number of emitters. The questions considering the limits on the size of the particle samples and their number statistics, applicability of possible nonclassicality in large ensembles for detection of related fundamental effects like quantum phase transitions, particle entanglement or its possible utilization for mesoscopic quantum computation still remain to be explored.

In this Letter, we present the experimental observation of nonclassical statistical properties of light emitted from large ensembles of single-photon emitters. We employ trapped ion crystals as a scalable source satisfying conditions for nonclassicality observability for large numbers of emitters. The recently proposed exactly measurable nonclassicality criterion, adjustable to the applied detection scheme [11], is used for witnessing the nonclassical character of emitted fluorescence. We measure the nonclassicality witness on emitted light for both pulsed and continuous laser excitation and verify the value of the nonclassicality threshold by conducting the same measurements with laser light.

Ensembles of large numbers of ions trapped in a Paul trap possess crucial advantages for initial proof-of-principle tests of nonclassicality [11,12]. First, average trapping lifetimes of single ions in our setup exceed several days, which allows for direct observation of emitted fluorescence, even for very large ion numbers [13–16]. Second, laser-cooled and trapped ion crystals can constitute isotopically pure samples of ions with lambda-type electronic level schemes. Because of their superb isolation from the environment and long trapping lifetimes, these systems have been employed for some of the pioneering tests of single-atom fluorescence nonclassicality [17,18] and more recently, led to the demonstration of single-photon sources with record-breaking single-photon content [19–22]. Furthermore, the typical interatomic distance of ions in the traps is limited to be much larger than the wavelengths of the involved optical transitions due to the Coulomb repulsion, which suggests that spontaneous buildup of collective effects can be neglected and ions can be treated as mutually independent emitters [23]. In addition, the ion-trapping apparatus allows for near perfect control of the number of emitters in the crystal due to easily repeatable, albeit probabilistic, ion-loading procedures.

In our measurements, we focus on reaching the maximal number of participating ions while still unambiguously demonstrating the nonclassicality of the emitted light field allowed by practical limits related to the ion-trapping and light collection apparatus. The simplified scheme of our experimental setup is shown in Fig. 1. The $^{40}Ca^+$ ion crystal is created by application of the trapping and Doppler cooling forces in the potential minimum of a linear Paul trap. The light scattered from ions is collected using a lens covering ≈2% of the full solid angle, with the radial position and focal point carefully optimized for maximizing the fluorescence detection efficiency from a single ion trapped in the trap center. See Supplemental Material, Sec. A [24] for more experimental details.

Nonclassical features of atomic fluorescence coming from trapped ion crystals are estimated by statistical analysis of the recorded time-tagged detection signal corresponding to the exact times of photon arrival at the two avalanche photodiodes (APDs) using the criteria from Lachman *et al.* [11] based on the bare estimation of the true photon detection probabilities. These criteria fundamentally differ from the measures that incorporate moments of photon distribution,

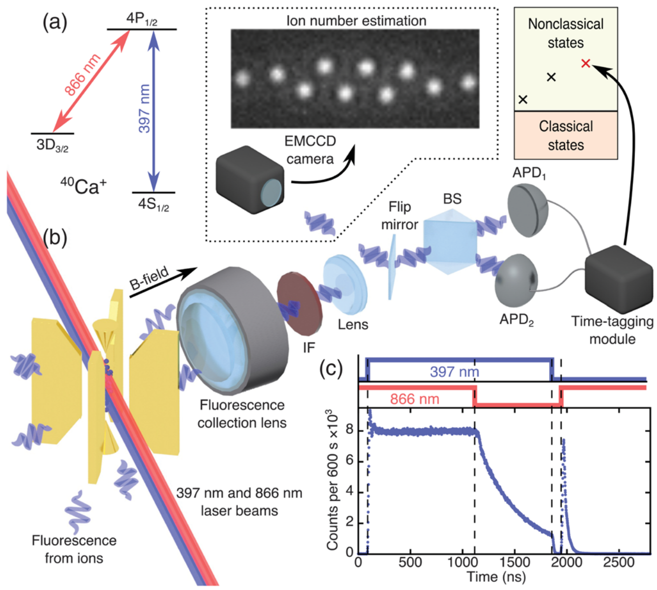

FIG. 1. The simplified scheme of the experimental setup and laser pulse sequence for generation and detection of nonclassical light. (a) The energy level scheme of the $^{40}Ca^+$ ion including employed transitions with their respective wavelengths. (b) An ensemble of ions is trapped in the linear Paul trap, and the 397 nm fluorescence emitted along the applied magnetic field direction (B field) is collected by a lens objective and separated from the 866 nm light by an optical interference filter (IF). The fluorescence is then directed towards the nonclassicality analyzing setup comprising a single beam-splitter (BS), a pair of avalanche photodiodes (APDs) and the time-tagging module. The trapped ion crystals can be imaged on an electron multiplying CCD (EMCCD) camera for the purposes of ion number estimation and optimization of trapping stability. (c) The generation of analyzed light from the trapped ion ensemble in the pulsed regime begins by the Doppler cooling period, in which both lasers are switched on. It is followed by the optical pumping stage, where the populations are shuffled to the metastable $3D_{3/2}$ state using only the 397 nm laser. The analyzed fluorescence at the $4P_{1/2} \leftrightarrow 4S_{1/2}$ transition is then generated by fast depopulation of the $3D_{3/2}$ state using the 866 nm laser pulse.

including commonly employed nonclassicality estimation methods based on measurements of the intensity correlation function $g^2(\tau)$. The estimation of $g^2(\tau)$ corresponds to the measurement of the photon number variance, which cannot be safely realized when using binary single-photon detectors for the observation of small nonclassicality from the large number of emitters. It generally requires the estimation of the whole photon number distribution. Although this is usually approximated by the probability of click and double click in the limit of small photon flux, such simplification is not safe in general and might become misleading, especially when the number of emitters, and thus, number of emitted photons, increases substantially [25,26].

The employed criterion [11] includes the real response of single-photon detectors and takes into account possible unequal quantum detection efficiencies. It is operationally derived from first principles by the construction of a linear functional depending on the probability of detecting no photon on both detectors, denoted P_{00}, and no photon on one particular detector, denoted P_0. The linear functional has the form

$$F_a(\rho) = P_0 + aP_{00}, \tag{1}$$

where a is a free parameter. If we consider a symmetrical detection scheme, optimizing $F_a(\rho)$ over all classical states $\rho = \int P(\alpha)|\alpha\rangle\langle\alpha|\mathrm{d}^2\alpha$ leads to the threshold function $F(a) = -1/(4a)$ which covers all classical states. Here $P(\alpha)$ is a density probability function. The nonclassicality condition requires a such that the detected probabilities satisfy $P_0 + aP_{00} > F(a)$. This a can be found if and only if $P_0 - \sqrt{P_{00}} > 0$. It thus allows for an unambiguous test of the nonclassicality of light even with a high mean number of photons, where the approximation of moments by probabilities of clicks is not generally safe because it can potentially imitate the nonclassical behavior [27]. The nonclassicality is then witnessed by estimation of the probabilities within a given time bin period and evaluation of the distance from the nonclassicality threshold

$$d = P_0 - \sqrt{P_{00}} > 0. \tag{2}$$

We note that the parameter d by no means gives a quantitative measure of nonclassicality; it is solely a suitable witness for nonclassical states from the large ensembles of emitters. The parameter d can be equivalently defined also in terms of the probability of a click P_s and a double click P_c; see Supplemental Material, Sec. D [24] for more details. However, the parameter d is better for understanding the experiments with many emitters. The value of d increases with the number of contributing single-photon emitters, while it is nonincreasing if noise, Poissonian or thermal, is added instead [11]. The $g^2(0)$ does not provide such information directly, as it converges to unity for the addition of both single-photon emitters and noise sources.

We measure the statistics of emitted fluorescence in both pulsed and continuous excitation regimes, which effectively represent different sources of radiation. In the continuous case, ions in the crystal emit fluorescence at random and mutually uncorrelated times, and, in principle, finite linewidth of the employed transition and laser light leakage can result in multiple emissions from a single ion within the same time bin. This is well illustrated by the decreased purity of the single photons emitted from single atoms, estimated from measurements of intensity correlation functions $g^2(0)$ in continuous schemes compared to pulsed sources, in which the multiphoton emission is typically prohibited by the optical pumping mechanism [19–22]. The pulsed driving is thus much more convenient for demonstration of the nonclassical emission from large atomic ensembles, provided that the rate of the photon emission given by the pulse sequence length can be kept comparably high. As can be seen in Figs. 1(a) and 1(c), we employ an effectively three-level energy structure of the $^{40}\mathrm{Ca}^+$ to minimize the multiphoton content in the given measurement time bin by optically pumping the atomic population to the metastable $3D_{3/2}$ level from where the photon emission is initiated by the 866 nm laser pulse. The optical pumping characteristic time is relatively long for our excitation parameters and crystal spatial extensions, and depending on the laser settings, about one fifth of emitters remain in the $4S_{1/2}$ level. The efficiency of the optical pumping to the $3D_{3/2}$ manifold is estimated from the observed fluorescence rates in the pulsed photon generation sequence; see the example in the Fig. 1(c). The residual average detected photon rate at the end of the 397 nm pulse is compared with the value of the photon rate during the Doppler cooling period, which gives a lower bound on the $3D_{3/2}$ population. The exact value can be then estimated by evaluating the steady state populations of the $3D_{3/2}$ state for given Doppler cooling laser excitation parameters. The single-photon detectors are gated with the gating time optimized to comprise most of the generated 397 nm light. The detailed description of the measured data processing can be found in Supplemental Material, Sec. B [24].

The evaluated value of witness d defined in Eq. (2) for trapped ion crystals containing from one to up to several hundreds of ions is plotted in Fig. 2(a). A clear violation of the nonclassicality condition by several error bars marking single standard deviation is observed for all ion numbers up to 275 in both continuous and pulsed regimes. The corresponding theoretical prediction of the parameter d in the pulsed regime has been evaluated and plotted by taking into account the measured distribution of detection efficiencies for ions in the given crystal. The theoretical prediction agrees well with the measured data, even without any free-fitting parameter and without taking into account experimental imperfections like the optical pumping spatial distribution or intensity fluctuations of the exciting lasers. In the presented simulation, we have considered the ion crystals with concentric shell structure supported by our observations. Details of the simulation can be found in Supplemental Material, Sec. C [24]. The ion storage lifetime and crystal stability can play a crucial role for the statistical properties of the emitted light. We have analyzed the rate of loss of ions in our setup in the regime where the cooling lasers frequencies are locked and the trap input radio-frequency power remains stable. The number of trapped ions was precisely counted before and after each photon-counting measurement, and there has been no observable loss of ions in the presented measurements. We note that only two out of 14 measurement runs were discarded due to loss of ions caused by cooling laser frequency instability. For comparison, the quality of our single-photon emitter-single ion has also been evaluated using the conventional measure based on the estimation of

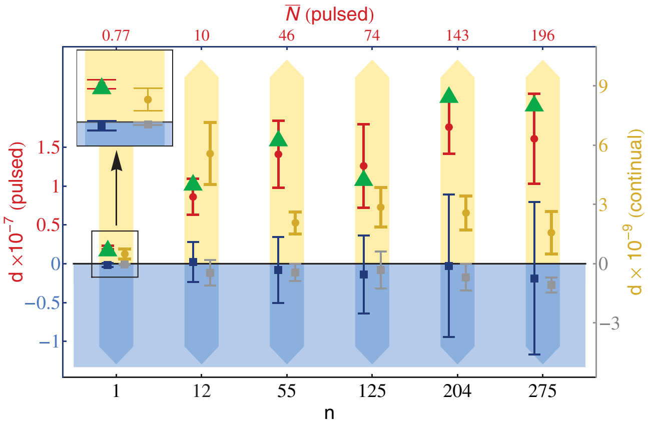

FIG. 2. The measured violation of nonclassicality as a function of the number of trapped single-photon emitters and the same measurements realized just with the laser light. All measured values of the witness d for trapped ion ensembles in pulsed (red circles) and continuous (yellow circles) regimes are provably in the nonclassical region given by $d > 0$. The green triangles correspond to the numerical simulation of the witness value in the pulsed regime; details of the simulation can be found in the Supplemental Material, Sec. C [24]. Estimated mean photon numbers per single emission pulse $\bar{N}$ in the pulsed regime are shown on the top axis. d for the 397 nm laser light scattered from the trap electrodes has been measured using the same detection scheme in both excitation regimes. The laser intensity has been set to reach the detection count rates corresponding to measurement on light from a given trapped ion ensemble. The dark blue and grey squares are measurements on the laser light in the pulsed and continuous regimes, respectively. All measurements corresponding to a given ion number are grouped in arrowlike shaded regions. Error bars correspond to one standard deviation.

the intensity correlation function $g^2(0)$, which gives $g^2(0) = 0.081$ in the continuous case for a 1 ns time bin and $g^2(0) = 0.032$ in the pulsed excitation regime, comparable to other realizations of single-photon sources with single trapped ions [19–22].

The internal atomic dynamics and multiphoton contributions make it difficult to theoretically predict the measured parameter d in continuous regime. The actual uncertainty in the emission time and possibility of multiphoton contributions from single atoms within finite time bins make this regime closer to a large class of optical sources with uncontrolled internal dynamics or partial coupling to environment [28], and the presented measurements will likely stimulate investigations of their statistical properties.

The technical limit on further increasing the number of emitters in our experiment is given by the effective detection volume of the employed optical detection setup. The photon detection efficiency falls rapidly for ions positioned in the radial direction from the collection lens focal point and less rapidly, but still considerably, along the lens symmetry axis. See Supplemental Material, Sec. A [24] for more details. The limiting radial size of the detection volume for our detection arrangement is 4.6 μm (FWHM). The detection efficiency variation at various distances from the lens focal point suggests that increasing the measured crystal size beyond hundreds of ions in our setup inevitably places some of them into regions with extremely small relative collection efficiency. The transition to small relative detection efficiencies is smooth in all three spatial directions, which brings in a technical question of how many emitters actually substantially contribute to the detected photon flux. This fuzziness in the number of contributing emitters is seen as a sign of dealing with a system that approaches the fragile borderline between the applicability of quantum and classical descriptions, at which the suitability of the discrete quantum description of some important physical variables, like number of emitters or total energy, naturally deteriorates. The mere technical limit of the employed optical detection setup can be eliminated by the use of optimized imaging configurations. We demonstrate further scalability of the nonclassicality measurements in similar setups by changing the lens configuration so that we decrease its overall magnification factor, which corresponds to an increase of the radial detection volume. We reach a radial field of view of approximately 20 μm without observing any substantial change of the absolute detection efficiency of an ion positioned on the optical axis. With this configuration, we have measured the positive distance $d = (9.48 \pm 3.93) \times 10^{-7}$ for 1500 ± 200 ions in an equal 5-hour-long experimental run. We note that this would correspond to 391 ions when considering only emitters contributing to the detected optical signal with relative efficiency higher than $\eta_{\max}/e$, where $\eta_{\max}$ is the overall detection efficiency for an ion positioned at the optical axis of the employed detection system and e is Euler's number.

The photon flux per single emission pulse at the input of the detection apparatus has been estimated for each measurement in the pulsed regime from the number of trapped ions n and measured finite efficiency of the optical pumping η_p as $\bar{N} = n \times \eta_p$. The highest mean photon number at the input of our detection apparatus is $\bar{N} = 196$ photons, which correspond to $n = 275$ ions and $\eta_p = 71\%$ optical pumping efficiency. To the best of our knowledge, this corresponds both to the largest ensemble of single-photon emitters and the largest photonic field for which nonclassical statistical properties have been demonstrated. Furthermore, as can be seen in Fig. 2(a), the relative uncertainty of the measurements of the distance d scales very favorably with the number of ions, which promises scalability of our measurements to much higher ion numbers, provided that the emitted light is collected efficiently.

Furthermore, there seems to be no fundamental limit on further substantial increase of any of these quantities in a similar experimental apparatus. We have simulated the emission of light from a crystal containing up to 10^5 ions. The simulation predicts scaling of the parameter d and uncertainty caused by the finite measurement with the size of the crystal. As already predicted in Ref. [11], the main limiting parameter for unambiguous detection of nonclassicality for stable ensembles of single-photon emitters is the measurement time. The detailed analysis presented in Supplemental Material, Sec. C [24] shows that the required experimental time does not grow substantially until the mean photon flux at photo detectors reaches several photons. The nonclassicality of stronger light would also be observable; however, it would require additional attenuation to reach the optimal photon flux for measuring nonclassical properties.

The presented measurements demonstrate the first unambiguous proof of the nonclassical character of light fields emitted from a large ensemble of single-photon emitters. We have shown that ensembles consisting of emitters which individually produce nonclassical light keep this statistical property when scaling up their size by at least two orders of magnitude by trapping and measuring for a range of ion numbers, from a single ion up to 275 ions. Moreover, the demonstrated nonclassicality measurements present robustness against many imperfections of individual sources. We have also verified several aspects of emission from the ensemble of single-photon sources compared to the emission from a single one. Most notably, the measured nonclassicality witness value in the pulsed regime grows or stays approximately constant with an increasing emitter number.

Our experimental test opens the possibility of searching for nonclassical light emission from recently developed ensembles of atomic [29,30] and solid-state emitters [9,10] and studying their internal dynamics from a new perspective [31–33]. It will allow their exploration before single emitters are isolated and further direct exploitation in quantum technology [34]. Furthermore, due to the large dependence of the ability to detect nonclassicality on the statistics of emitters and emitted light mode-structure stability, the presented scheme can be easily applied for detection of phase transitions from solid to gas or plasma phase [33]. The presented demonstration can be directly extended in the future to ion numbers beyond thousands by employing optimized optical fluorescence collection schemes, and later, to observations of emissions going toward the Fock states of indistinguishable photons from atoms inside a cavity [35,36]. It substantially shifts the range of energies in which observable manifestations of discrete quantum features of light and matter should be anticipated and thus will likely trigger the construction of truly macroscopic and intense sources of quantum light.

In the course of preparing our Letter, we became aware of another experiment studying nonclassicality from ensembles of single-photon emitters [37]. It demonstrates the applicability of the presented proof-of-principle methodology for studies of emission from clusters of nitrogen-vacancy centers in diamond and its robustness against realistic sources of noise.

The work reported here has been supported by Grant No. GB14-36681G of the Czech Science Foundation. We acknowledge the kind technological support from the group of Rainer Blatt.

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