Direct experimental observation of nonclassicality in ensembles of single-photon emitters

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(Received 19 May 2017; revised manuscript received 7 November 2017; published 27 November 2017)

In this work we experimentally demonstrate a recently proposed criterion addressed to detect nonclassical behavior in the fluorescence emission of ensembles of single-photon emitters. In particular, we apply the method to study clusters of nitrogen-vacancy centers in diamond characterized with single-photon-sensitive confocal microscopy. Theoretical considerations on the behavior of the parameter at any arbitrary order in the presence of Poissonian noise are presented and, finally, the opportunity of detecting manifold coincidences is discussed.

DOI: 10.1103/PhysRevB.96.195209

I. INTRODUCTION

One of the most debated issues in quantum mechanics is related to understanding the boundary separating the counter-intuitive behavior of the systems governed by the quantum laws from the classical, familiar properties of the macroscopical systems. This transition also manifests itself in the realm of optics [1] where, even if undoubtedly the radiation emitted by any possible source of light is indeed composed by an ensemble of individual photons, the properties of classical sources differ consistently from those of nonclassical ones. In particular, single-photon sources (SPSs) have found many experimental and reliable realizations in systems such as heralded sources based on parametric down-conversion [2–9], quantum dots [10,11], trapped ions [12], molecules [13], and color centers in diamond [14–21]. Since nonclassical optical states have now become a fundamental resource for quantum technology [22,23], the determination of nonclassicality [24] for a state is not only important for studies concerning boundaries from quantum to classical world, but also represents an important tool for quantifying such resources. Vast literature exists on the characterization of SPSs [25]. Most of the techniques rely on the sampling of the second-order autocorrelation function (or Glauber function)

$$g^{(2)}(\tau=0)=\left.\frac{\langle I(t)I(t+\tau)\rangle}{\langle I(t)\rangle\langle I(t+\tau)\rangle}\right|_{\tau=0}, \qquad (1)$$

whose value is never smaller than 1 for classical light, while it is lower than 1 for sub-Poissonian light, and in particular vanishes for single-photon states, where $g^{(2)}(0)=0$ is expected in the ideal case. This quantity has been shown to be substantially equivalent to the parameter α introduced by Grangier *et al.* [26] (and throughout the paper we will refer to this parameter as $g^{(2)}$ without distinction), which is experimentally measured as the ratio between the coincidence probability at the ouput of a Hanbury Brown and Twiss interferometer (HBT) [27], typically a 50:50 beam splitter connected to two non-photon-number-resolving (non-PNR) detectors, and the product of the click probabilities at the two detectors [22]. This parameter can be generalized to account for the statistical properties of N-fold coincidence events at the outputs of detector-tree apparatuses and several techniques for the reconstruction of optical states as well as quantum enhanced imaging techniques are allowed by the experimental sampling of $g^{(N)}$ functions [28–44]. Unfortunately, the amount of background light can affect the measurement, leading to a camouflage of the quantum characteristics due to noise. More specifically, in practical cases, when sampling $g^{(2)}(0)$ to characterize single emitters it is not possible to distinguish between the true quantum signal and background light contribution and, in extreme cases, one is not able to detect a single emitter drowned in dominant noise bath. Recently a novel criterion allowing one to reveal no-classical light from large numbers of independent SPSs has been proposed [45]. According to the theoretical predictions, an experimental implementation of this criterion would be extremely advantageous not only because it would allow one to spot nonclassical signatures in the emission of clusters of emitters, but also because it can be shown that this technique is extremely robust in the presence of Poissonian noise, the parameter under test being absolutely independent from this kind of noise contribution (even when it is dominant).

In this work we experimentally apply the criterion [45] to directly detect nonclassical emission from ensembles of SPSs based on nitrogen-vacancy (NV) centers in nanodiamond observed by means of a confocal microscope coupled to four non-PNR single-photon detectors in a detector-tree configuration. Although the reported methodology can be generally extended to a broad range of different physical systems, NV defects in diamond have been elected to benchmark this new criterion. This choice is motivated by the high relevance of this physical system in quantum optics due to its appealing spin-dependent transition structure, as widely demonstrated by a broad range of works on the subject in recent years [46]. Moreover, the choice is motivated by the fact that generally the quantum-optical characterization of individual photon emitters in solid-state systems (such as NV and other color centers in diamond) is significantly affected by complex issues in the correct assessment of sources of background luminescence in confocal microscopy, such as nearby defects, scattered light, diffused fluorescence from extended defects, ambient light, etc. [14,17], which are not always easily manageable, or duly taken into account.

II. THEORETICAL MODEL

In general, the system considered here is an ensemble of M single-photon emitters, each coupled to the detection

