Beating the Abbe Diffraction Limit in Confocal Microscopy via Nonclassical Photon Statistics

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We experimentally demonstrate quantum enhanced resolution in confocal fluorescence microscopy exploiting the nonclassical photon statistics of single nitrogen-vacancy color centers in diamond. By developing a general model of superresolution based on the direct sampling of the kth-order autocorrelation function of the photoluminescence signal, we show the possibility to resolve, in principle, arbitrarily close emitting centers.

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In the last decade, measurement techniques enhanced by using peculiar properties of quantum light [1,2] have been successfully demonstrated in several remarkable real application scenarios, for example, interferometric measurements aimed to reveal gravitational waves and the quantum gravity effect [3,4], biological particle tracking [5], phase contrast microscopy [6], and imaging [7,8]. Very recently, a novel technique to beat the diffraction limit in microscopy that relies on the antibunching behavior of photons emitted by single fluophores has been proposed [9], and realized in wide field microscopy [10] by using an EMCCD camera.

The maximum obtainable imaging resolution in classical far-field fluorescence microscopy, according to the Abbe diffraction limit, is $R \simeq 0.61\lambda/\mathrm{NA}$, where λ is the wavelength of the light and NA is the numerical aperture of the objective. This restricts the current capability of precisely measuring the position of very small objects such as single photon emitters (color centers, quantum dots, etc.) [11–19], limiting their potential exploitation in the frame quantum technology [20,21]. In general, the research of methods to obtain a microscopy resolution below the diffraction limit is a topic of the utmost interest [22–29] that could provide dramatic improvement in the observation of several systems spanning from quantum dots [30] to living cells [31–34]. As a notable example, in several entanglement-related experiments using strongly coupled single photon emitters it is of the utmost importance to measure their positions with the highest spatial resolution [35]. In principle, this limitation can be overcome by recently developed microscopy techniques such as stimulated emission depletion (STED) and ground state depletion (GSD) [36,37]. Nevertheless, even if they have been demonstrated effectively able to provide superresolved imaging in many specific applications, among which are color centers in diamond [38], they are characterized by rather specific experimental requirements (dual laser excitation system, availability of luminescence quenching mechanisms by stimulated emission, nontrivial shaping of the quenching beam, high power). Furthermore, these techniques are not suitable in applications in which the fluorescence is not optically induced [39,40], so that new methods are required for those applications.

Inspired by the works in [9], in this Letter we develop a comprehensive theory of superresolution imaging of clusters of single photon emitters based on high order Glauber correlation functions $g^{(k)}(t=0)$. Our theory discloses the unexpected possibility of approaching an arbitrary resolution just by measuring the spatial map of the correlation up to k_0th order when it is reasonable to assume $g^{(k)}=0$ for $k>k_0$. For example, two arbitrarily closed emitters can be, in principle, separated just by measuring $g^{(2)}$ being of course $g^{(3)}=0$. Then, it confirms the indication of [9] that a fair $1/\sqrt{k}$ improvement of resolution can be obtained with the measurement of $g^{(k)}$, if no further information is available. We experimentally test the theory of quantum superresolution in the significant case of confocal microscopy for the first time, considering clusters of few NV centers in artificial diamond grown by chemical vapor deposition and using a detector-tree of commercial (non-photon-number-resolving) single photon detectors [18,41]. We demonstrate a resolution increase by sampling the $g^{(2)}$ of the signal, and a further improvement by measuring $g^{(3)}$. Furthermore, we show that just by considering the contribution of higher powers of $g^{(2)}$, when only two centers are relevant (as certified by $g^{(3)}=0$), larger improvement in the resolution can be obtained, as predicted by the theory. This technique appears particularly valuable since the sampling of $g^{(2)}$ is a widely used and

