

Quantum Statistical Imaging of Particles without Restriction of the Diffraction Limit

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A quantum measurement method based on the quantum nature of antibunching photon emission has been developed to detect single particles without the restriction of the diffraction limit. By simultaneously counting the single-photon and two-photon signals with fluorescence microscopy, the images of nearby nitrogen-vacancy centers in diamond at a distance of 8.5 ± 2.4 nm have been successfully reconstructed. Also their axes information was optically obtained. This quantum statistical imaging technique, with a simple experimental setup, can also be easily generalized in the measuring and distinguishing of other physical properties with any overlapping, which shows high potential in future image and study of coupled quantum systems for quantum information techniques.

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The measurement of physical quantities is not only a major goal but also an active impulsion for scientific research. In particular, the imaging of nearby particles is important for modern science [1,2]. The precision with which two nearby particles can be resolved is classically restricted by the optical diffraction limit. Imaging methods that used distinguishing information based on the photons emitted from different particles have been proposed to achieve precision beyond the diffraction limit [3–10]. When the emitted photons have the same properties, distinguishing nearby particles which are separated by distances much less than the diffraction limit is difficult.

Recently, phenomena from quantum mechanics have been used to improve the measurement and have been applied to some special purposes which cannot be performed by classical method. Such quantum techniques are being applied to enhance the precision of measurements beyond the classical limit [11,12]. However, many quantum-based protocols to improve the measurement used the quantum entanglement. They are fragile because of quantum decoherence [13–15]. For practical purpose, stable quantum phenomena should be applied in the measurement. The temporal fluctuation [9] and sub-Poissonian statistics [10] have been applied to enhance the imaging resolution by $\sqrt{N}$ with an N th-order process. Here, we have developed a quantum statistical imaging (QSI) method to detect single particles without the restriction of the diffraction limit. In the quantum regime, the situation for particle imaging is different because each particle emits only one photon and shows the single-photon antibunching effect [16]. By detecting the photon coincident counts, the particles can be imaged and resolved even when they are almost completely overlapping and the emitted photons are identical. Here, nitrogen-vacancy center (NVC) in diamond was used in this experimental demonstration. Single NVC has shown its good quality as a single-photon source [17,18]. When two NVCs are close to each other, i.e., within tens of nanometers, the strong dipole-dipole

interaction can be applied in quantum information techniques [19,20]. Diamond nanocrystals with NVCs have been successfully used to image biological processes [21,22]. The ability to image and distinguish two nearby NVCs is becoming increasingly important in physics [19,20,23] and biology [21,22].

NVCs are usually detected by scanning confocal optical fluorescence microscopy [24], in which the single-photon intensity of spontaneous emission is measured to characterize the optical images of the centers. The spontaneous emission from the NVCs, which were fabricated by nitrogen ion implantation, is collected into a single-mode fiber. The collected photons are then split into two paths by a fiber beam splitter with a transmissivity of T and a reflectivity of R ($T + R = 1$) to form a Hanbury Brown–Twiss interferometer [25]. Finally, the separated beams are detected with two single-photon detectors. When two NVCs (A and B) are imaged, the single-photon intensity at position (x, y) from the single-photon detectors D_1 and D_2 can be expressed as

$$\langle I_1^{D1}(x, y) \rangle = T[\langle I_A(x, y) \rangle + \langle I_B(x, y) \rangle],$$

$$\langle I_1^{D2}(x, y) \rangle = R[\langle I_A(x, y) \rangle + \langle I_B(x, y) \rangle],$$

where $\langle I_A(x, y) \rangle$ and $\langle I_B(x, y) \rangle$ are the single-photon rates from NVCs A and B , respectively. Therefore, the single photons from the two NVCs are

$$\begin{aligned} \langle I_1(x, y) \rangle &= \langle I_1^{D1}(x, y) \rangle + \langle I_1^{D2}(x, y) \rangle \\ &= \langle I_A(x, y) \rangle + \langle I_B(x, y) \rangle. \end{aligned} \quad (1)$$

When the distance between the two NVCs is within the optical diffraction limit, $\langle I_A(x, y) \rangle$ and $\langle I_B(x, y) \rangle$ are overlapping and hardly distinguishable from the single-photon intensity $\langle I_1(x, y) \rangle$. If two-photon coincident counts are measured, then the two photons must come from two NVCs and never from the same NVC because a single NVC only emits one photon. This attribute demonstrates a genuine quantum characteristic, namely, the photon

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marked as “+” or “×,” only one NVC is effectively excited as another NVC is out of the exciton spot. Therefore, $g_c^{(2)}(0)$ is close to zero as a single NVC. From marked positions “+” to “×” in Fig. 2(a), it is clear to see the variation from single to double NVCs.

For the first pair, the single-photon intensity [$\langle I_1(x, y) \rangle$] and two-photon intensity [$\langle I_2(x, y) \rangle$] were recorded at the same time [Figs. 3(a) and 3(b)]. In Fig. 3(b), the image of the two-photon intensity has a narrower width in the overlapping region of the two NVCs. However, these images do not provide spatially resolved images of the two NV centers. The image of a single NVC should have a single peak and a continuous envelope. Using $\langle I_1(x, y) \rangle$ and $\langle I_2(x, y) \rangle$, the photon intensities $\langle I_A(x, y) \rangle$ and $\langle I_B(x, y) \rangle$ of the two particles can be obtained by solving Eqs. (1) and (2). The images of the two particles, I_A and I_B , were reconstructed and are shown in Figs. 3(c) and 3(d). By fitting the data, the distance between the NVCs was determined to be 366.1 ± 2.8 nm, which is at the edge of the diffraction limit. Figure 3(e) shows a 3D image of the two nearby NVCs, and the overlapping can be observed.

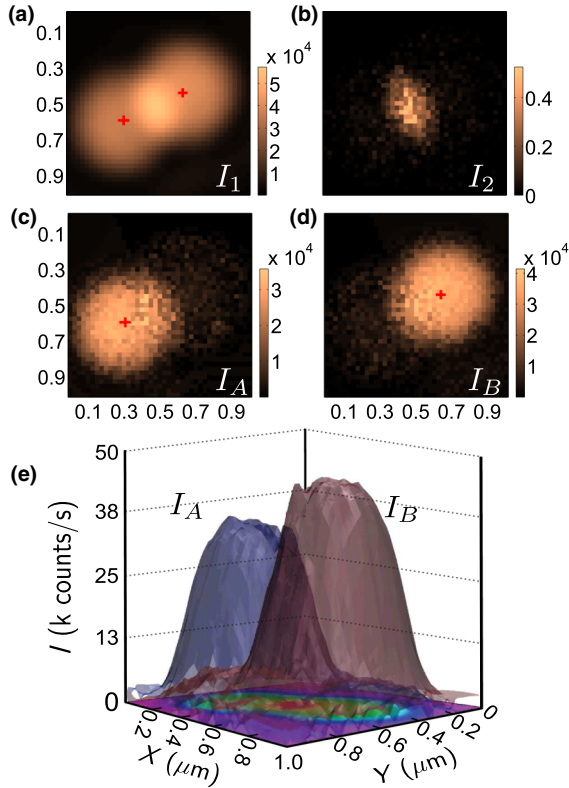


FIG. 3 (color online). Optical images of two single NVCs at a distance of 366.1 ± 2.8 nm. (a),(b) I_1 and I_2 for the single-photon and two-photon counts. (c),(d) Images of each NVC (I_A , I_B), respectively. The red crosses mark the positions of the NVCs with an uncertainty of $1/20$ the length of each cross. The positions were obtained using a 2D Gaussian fitting of I_A or I_B . (e) 3D image of the two NVCs.

Another pair of NVCs with a much smaller separation was also measured and distinguished. The confocal scanning image, the result of photon autocorrelation measurement, and the spectrum show that there are two NVCs (see Fig. 2S in the Supplemental Material [29]). The single-photon and two-photon images are shown in Figs. 4(a) and 4(b). From the single-photon intensity, the two NVCs are well overlapping and cannot be distinguished. Using the QSI method, their images, I_A and I_B , can be obtained, as shown in Figs. 4(c) and 4(d). The distance between the centers was determined to be 8.5 ± 2.4 nm by fitting I_A and I_B . This distance is much smaller than the diffraction limit, and the NVCs cannot be readily distinguished by the classical method [19]. Here, the resolution (see Supplemental Material for details [29]) was determined by the number of recorded photons and the experimental setup. In the present measurement, the error is about 0.9 nm with 10^5 coincident photon counts and the setup repeat resolution for the x or y axis is about 0.7 nm.

In addition to imaging nearby particles, such a quantum measurement method can be easily generalized to measure and distinguish other properties even with high overlapping. For example, the axes of the NVCs can be measured. The spontaneous emission rates vary with the polarization of the pump beam according to different axes of NVCs [30,31].

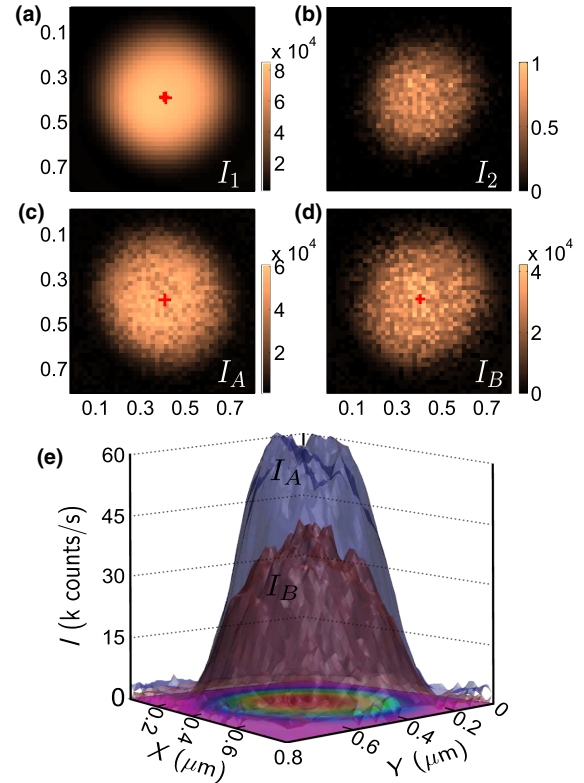


FIG. 4 (color online). Optical imaging of two single NVCs with a small separation. The images are organized as in Fig. 3. The distance between the NVCs was determined to be 8.5 ± 2.4 nm by fitting I_A and I_B .

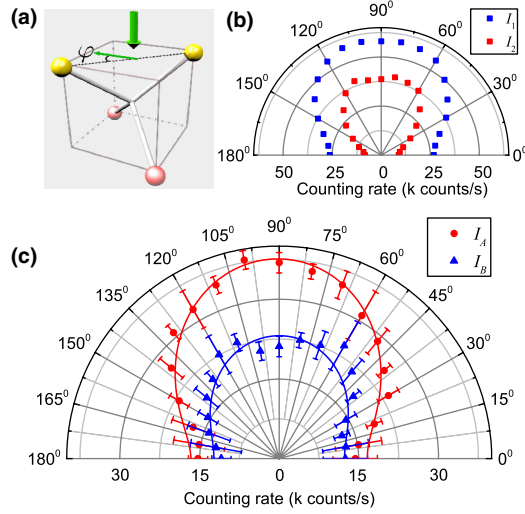


FIG. 5 (color online). (a) The schematic of two sets of axes for [100]-oriented NVC sample according to the polarization of pump beam. They are in the plane of $\varphi = 0^\circ$ or $\varphi = 90^\circ$. (b) Single-photon and two-photon emission intensity versus the polarization angle of pump beam. The two-photon intensity was amplified by 5×10^3 for view. (c) Separated intensities of two NVCs versus the polarization angle of pump beam.

With polarized optical pump for our [100]-oriented sample, the number of possible orientations of a given center is reduced from four to two, which are in the plane of $\varphi = 0^\circ$ or $\varphi = 90^\circ$, as shown in Fig. 5(a). Here, with QSI, the axes of the pair of NVCs at the distance of 8.5 nm have been obtained. With different polarized pump beam, the single-photon and two-photon counts are shown in Fig. 5(b). Simply, the emission intensity of each NVC with the angle of polarization can be obtained in Fig. 5(c). Correspondingly, the axes of the two NVCs are the same in the plane of $\varphi = 0^\circ$. The data can be fitted with $I_{A(B)} = \alpha_{A(B)} + \beta_{A(B)} \cos^2 \varphi$ [31] with $\alpha_{A(B)} = 37.5 \pm 0.4 (23.1 \pm 0.5)$ and $\beta_{A(B)} = -21.0 \pm 0.8 (-10.8 \pm 0.9)$. The small dip at $\varphi = 90^\circ$ for NV_B may come from its depth in the diamond [30].

In addition to the demonstrated symmetric envelopes of the photons from NVCs, the QSI method can be applied to detect and distinguish images with other continuous envelopes. In the proposals from Refs. [9,10], the resolution improvement is acceptable for Gaussian distribution. For other types of distribution, these methods will encounter some difficulties. Also, some other conditions should be met to give the enhancement in Ref. [9], which are not required by the QSI method. Furthermore, the QSI method can be used to detect other particles and distinguish other degrees of freedom, such as lifetime, frequency, and polarization. Because the photons from each particle can be recorded simultaneously and distinguished without decoupling nearby particles, the QSI method can be used to detect their separate dynamics, as well as the coupling between them. Also, the experimental setup for QSI is

simple. It does not need complicated pump beams [19,23] or the assistance of other control systems [19,20]. The limitation of the application and generalization of QSI may come from the photon collection. In the current experiment, the single-photon counts were about 20 K/s and the total detection efficiency was about 0.15%, including the loss at the collection objective (97%), pass loss (90%), and the loss at the detector (50%). However, in principle, all of these performances can be much improved. The collection efficiency can be enhanced by a factor of 8 with structured interface [32]. The loss from the beam splitter can be removed with additional detectors [15]. The loss at the detector can be overcome with high quantum efficiency. If a pulsed laser is used, there is no need for the 2 ns gate and all the photons within its lifetime can be collected, which is about a sixfold enhancement in the two-photon counts. Along with other low loss filters, the total two-photon detection rate can be improved by over 3 orders of magnitude. For the image of NVCs, the sample was moved at 40×40 locations (pixels) and photon counts were collected at each location. A gated CCD can be used to give additional enhancement in the data collection. With the above improved techniques, the total data collection time will be less than 1 min for the two NVCs with the same resolution, compared to the current 50 h. It is very promising in the scalable application for multipartite measurement and the detection of particles which emit small numbers of photons because of bleaching or blinking. In those cases, the resolution is limited by the total photons. However, it will still have very good resolution. For example, 10^5 photons can offer the subnanometer resolution.

In summary, a QSI method was demonstrated based on the unique quantum behavior of antibunching emission photons. Two well-overlapping NVCs can be spatially resolved. Also, the axes of the two NVCs are measured even though their distance is within 8.5 nm. With high order of coincident multiphoton measurements, additional single NVCs can be imaged and distinguished. The scalable QSI with high order coincidence counts will be applied in the multipartite interaction for quantum information techniques and modern physics.

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