Experimental demonstration of random walk by probability chaos using single photons

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In our former work, we demonstrated chaotic oscillation induced in a nanoscale system consisting of quantum dots between which energy transfer occurs via optical near-field interactions. It is intriguing that the chaotic behavior is associated with probability. Indeed, we previously examined such oscillating probabilities via diffusivity analysis by constructing random walkers driven by chaotically driven bias. Herein, we experimentally implemented the concept of probability chaos using a single-photon source that was chaotically modulated by an electro-optical modulator that directly yielded random walkers via single-photon observations. An evident signature was observed in the ensemble average of the time-averaged mean square displacement. © 2020 The Japan Society of Applied Physics

Energy transfer based on optical near-field interactions has been intensively studied both theoretically1) and experimentally.2,3) The optical excitation generated in the energy level of the smaller quantum dot, denoted by S1 in Fig. 1(a), can be transferred to the upper energy level of the larger dot L2 through optical near-field interactions.4) The excitation on L2 could then be shifted to the lower energy level L1 via energy dissipation. Such near-field processes have been utilized in lighting and energy applications.5) Additionally, fundamental studies toward computing and intelligent functions such as solution searching,6) decision making,7) and computing8) are emerging to benefit from the novel optical mechanisms inherent in the subwavelength scale. In the context of functional systems, the generation of periodic signals is one of the most fundamental aspects of the functional systems observed in nature, such as heart beats, and artificially constructed devices, such as clock signals for microprocessors. Indeed, Naruse et al. theoretically demonstrated optical pulsation using energy transfer among quantum dots interacted via near-field interactions9) by incorporating a delay mechanism as schematically indicated by \( \Delta \) in Fig. 1(a).

Furthermore, combined with additional external delay, indicated by \( \Delta_C \) in Fig. 1(a), chaotic oscillations and random number generations have been demonstrated.10) The solid curves in Figs. 1(b,i), 1(b,ii), 1(b,iii), and 1(b,iv) show the time evolution of the population associated with the lower energy level of the larger quantum dot L2 when the coupling strength \( \alpha_C \) between the external delay system and the original energy transfer system is given by \( 30 \times 10^{-3}, 180 \times 10^{-3}, 182 \times 10^{-3}, \) and \( 207 \times 10^{-3} \), respectively.10) In Fig. 1(b), periodically oscillating evolution of the population in (i), chaotically evolving populations in (ii) and (iii), and a quasi-periodically evolving population in (iv) are evident. Let \( \alpha_C \) be a control parameter, and the red circular and the blue x marks in Fig. 1(c) represent local maxima and local minima of the populations, respectively. Bifurcations are clearly observed, which were adapted from Fig. 3 of our former work.10)

In addition to the nanoscale implementation of such oscillatory behavior, it is intriguing that the observed time evolutions are associated with the elements of a density matrix, meaning that what is oscillating is probability. To examine the nature of such oscillatory probability, in the study in Ref. 11, we conducted a diffusivity analysis by constructing random walkers driven by dynamically changing bias supplied by the time evolution of the populations calculated from the quantum dot systems involving near-field interactions and time delay. We examined the ensemble average of the time-averaged mean square displacement (ETMSD) of the random walkers, defined as

\[
\text{ETMSD}(\Delta) = \left\langle \frac{1}{T - \Delta} \sum_{t=1}^{T-\Delta} \Delta x_s(t; \Delta)^2 \right\rangle_s
\]

where \( \Delta x_s(t; \Delta) = x_s(t + \Delta) - x_s(t) \) denotes the displacement of the walker and \( \langle \cdot \rangle_s \) denotes an ensemble average. One of the major results therein was that the ETMSD underwent large deviations from linear scaling in short time intervals in the case of chaotic oscillations, exhibiting a super-diffusion-like behavior. Such large diffusivity implies benefits in applications such as decision making; for example, the chaotic time series generated by a semiconductor laser provide efficient solutions to the multi-armed bandit problem.12)

Based on these earlier works, here we experimentally implemented the concept of dynamically changing probability, including chaotic changes, by using a single-photon source modulated by an external electrooptic modulator. The single-photon was then subjected to a polarization beam splitter (PBS) and detected by either of the two avalanche photodiodes (APDs) corresponding to horizontal or vertical polarization. As explained in detail below, the identity of the APD corresponds directly to the direction of the random walker. Consequently, the diffusivity is evaluated directly from the photon measurement sequences. Although the experiment was a scaled-up, proof-of-concept model of the genuine nanoscale oscillators, the experimental observations clearly validate the concept of oscillating probability, paving the way for future nanoscale systems.

The experimental setup is schematically illustrated in Fig. 2. A single-photon is emitted from a single individual nitrogen-vacancy (NV) color center,13) which has a broadband emission spectrum in the visible range (640–700 nm),14–16) excited by a...
Fig. 1. (Color online) Chaos in nano-optical pulser. (a) When optical excitation transfer via near-field interactions between smaller and larger quantum dot involves a time delay $\Delta$, optical pulsation becomes possible. Combined with additional external delay $\Delta_C$, irregular dynamics appears, including chaos. (b) Evolutions of populations from the system: (i) periodic, (ii) quasiperiodic, and (iii) and (iv) chaotic. (c) With the change in control parameter $\alpha_C$, a clear bifurcation diagram is observed. The images were adapted from Naruse et al., Sci. Rep. 4, 6039 (2014). Copyright 2014 Author(s), licensed under a Creative Commons Attribution 4.0 License.
green laser (CNI Lasers, diode-pumped solid-state laser; Wavelength: 532 nm). After passing through a polarizer (denoted by Pol in Fig. 2) and a zero-order half-wave plate ($\lambda/2$), the emitted photon impinges on an electro-optical phase modulator (Thorlabs, EO-PM-NR-C1) driven by an external system through an amplifier. The half-wave voltage of the modulator was 205 V at 633 nm. The modulated signal is subjected to the PBS, followed by single-photon detection by either APDH or APDV (Excelitas Technologies, SPCM-AQRH-16-FC), which correspond to horizontal and vertical polarization, respectively. The signal is then sent to a time-correlated single-photon-counting system (PicoQuant, PicoHarp 300).

Assume that there is no modulation by the modulator and that the polarization of the input single-photon is 45° with respect to the horizontal axis; the photon is detected by APDH or APDV with 50:50 probability. Next, the single photons are modulated by a periodically oscillating signal, which is shown in Fig. 1(b,i), using the electro-optical phase modulator. In this study, the time scale of the nanoscale simulations in Fig. 1(b) was stretched by about a factor of $10^7$ in the driving signal in the experiment taking into account the experimentally available single-photon rate (which is on the order of $10^4$ s$^{-1}$). The red and green traces in Fig. 3(a) show the number of photons per 2 ms duration time bin detected by APDH and APDV, respectively. The total duration of the measurement was about 12.7 s. Figure 3(b) is a magnified view of Fig. 3(a) corresponding to the initial 1 s. From these images, we can observe that the polarization of the incoming single photons is modulated by the periodic signals, while photons are detected simultaneously at either APDH or APDV, especially when the periodical modulation signal crosses the zero point. That is, the probabilistic attributes of single photons are also observed.

Precisely, the photons were detected as schematically shown in Fig. 3(c), where single-photon detection events are depicted by red and green x marks, which correspond to detection by APDH and APDV, respectively. In discussing the diffusivity induced by chaotically driven bias, we constructed a random walker directly from the single-photon detection events using the APDs. Specifically, the position of the random walker was updated by

$$x(t + 1) = \begin{cases} x(t) + 1 & \text{if photon is detected by APDH}, \\ x(t) - 1 & \text{if photon is detected by APDV}. \end{cases}$$

assuming the initial position of the random walker to be zero $[x(0) = 0]$. Here, the time $t$ is updated when a single-photon is detected by either of the detectors.

To give a precise description, we subtracted the single-photon detection events from the observed photon sequences; this was done to avoid the trend of an imbalance between APDH and APDV originating from the minute differences in sensitivity, optical misalignments, etc, of the photon detections.

We prepared five kinds of modulation signal trains subjected to the optical phase modulator based on the numerical study concerning near-field-mediated energy transfer in Ref. 10:

- **(1) Constant** Constant value: The polarization of single photons is maintained at about 45° with respect to the horizontal.
- **(2) Periodic** A periodic signal specified by $\alpha_C = 30 \times 10^{-3}$, which is referred to as “30” hereafter, calculated in the near-field energy transfer calculations. [See Fig. 1(b,i).]
- **(3) Quasiperiodic** A quasiperiodic signal specified by $\alpha_C = 207 \times 10^{-3}$ (called “207”). [See Fig. 1(b,iv).]
- **(4) Chaos 1** A chaotic signal specified by $\alpha_C = 180 \times 10^{-3}$ (called “180”). [See Fig. 1(b,ii).]
- **(5) Chaos 2** A chaotic signal specified by $\alpha_C = 182 \times 10^{-3}$ (called “182”). [See Fig. 1(b,iii).]

It is worth noting that the populations shown in Fig. 1(b) only represent the initial 50 ns evolutions. The number of data points in the sequences (2) to (5) was 900,000 with a time interval of 1 ps after omitting the initial 100 ns to avoid transient periods. For each of the signal trains [(1)–(5)], the
measurements were conducted 10 times. From each measure-
ment, a walker $x_i(t)$ was generated based on the rule given
by Eq. (2), followed by calculating the time-averaged mean
square displacement, which is the inner component of Eq. (2)
given by
\[
\text{TAMSD}_t(\Delta) = \frac{1}{T - \Delta} \sum_{t=1}^{T-\Delta} [x_i(t; \Delta)]^2.
\]
whose ensemble average yields ETMSD: \( \text{ETMSD}(\Delta) = \langle \text{TAMSD}_t(\Delta) \rangle \).

As is well known, when a random walker moves toward
the plus and minus directions with probabilities $p$ and $q$,
respectively, the mean displacement is given by \( \langle x(t) \rangle = (p - q)t \), while the variance is given by
\( \langle (x(t) - \langle x(t) \rangle)^2 \rangle = 4pqt \). Therefore, if the probability of
single-photon detection by APD$_H$ and APD$_V$ is 50:50, the
mean square displacement at time $t$ is equal to $t$ since
$p = q = 1/2$, which is called normal diffusion. The black
solid line in Fig. 4(a) depicts normal diffusion, meaning that
the ETMSD is equal to the time difference $\tau$. The magenta
marks in Fig. 4(a) show the ETMSD when the modulation
has a constant value [(1) Constant], which almost perfectly
agrees with the normal diffusion. The red circular marks
correspond to the case with periodically oscillating modula-
tion [(2) Periodic], where the ETMSD is clearly modulated in
a sinusoidal manner. The green, blue, and cyan marks
correspond to modulation by [(3) Quasiperiodic], [(4)
Chaos 1], and [(5) Chaos 2], respectively, which quickly
deviate from the normal diffusion. Especially with chaotic
modulation, the ETMSD exceeds 250 at approximately
$\tau = 9$ s, which is larger than the normal diffusion by a factor
of approximately 30. As was discussed in Ref. 11, the large
deviation from the normal diffusion with chaotic modulation
concerns the regions when the time difference is not large.
Indeed, the ETMSD decreases after approximately $\tau = 9$ s.
Nevertheless, the large departure from the normal when $\tau$
is small is an interesting feature, which has been theoretically
examined using the notion of generalized random walk that
covers Lévy walk.\textsuperscript{11} Intuitively, the persistence time of a
walker walking to the same direction, by the chaotically
modulated bias, provides the deviation from the normal. The
ETMSD curve by (2) periodic modulation exhibits mono-
tonously modulated depending on the value of $\tau$. At present,
we do not have full understanding of the underlining
mechanism of this particular observation, which could be a topic for future study.

Figure 4(b) depicts scatter plots of the ensemble average of the position of the walker at time step $t$, $\langle x(t) \rangle$, versus that of time step $t + D$, $\langle x(t + D) \rangle$, with $D$ being 10 000 considering the five kinds of modulation signals. The blue and cyan marks are based on the experimental results using chaotic modulations (4) Chaos 1 and (5) Chaos 2, respectively, whereas the magenta, red, and green marks correspond to (1) constant, (2) periodic, and (3) quasiperiodic signals. It can be observed that, with chaotically modulated single photons, the walker travels a larger area in the phase diagram of $\langle x(t) \rangle$, $\langle x(t + D) \rangle$ whereas its diffusing space is rather limited with the other modulations. These observations are consistent with the numerical studies described in Ref. 11.

To summarize, we experimentally examined the concept of dynamically changing probabilistic behavior by combining single photons emitted from a NV center in a nanodiamond and an electro-optical phase modulator. The single-photon measurements made by either of the two photodetectors corresponding to horizontally and vertically polarized light directly generated random walkers, whose diffusivities were analyzed using the mean square displacement as well as by examining the trajectories of the walkers in a phase space. Super-diffusion-like behavior was observed on a short time scale by chaotic modulation of single photons. The results agree well with the theoretical and numerical findings reported in Ref. 11. Although this is a scaled-up, proof-of-concept study, it is noteworthy that large diffusivity was experimentally achievable using a
rather simple experimental setup, motivating future implementation to nanoscale optical energy transfer, as originally discussed in Ref. 10. A relevant theoretical examination of a nano-scale oscillator is the principle of superradiance proposed by Shojiguchi et al.17) The experimental realization would, for either method, require breakthroughs in device and material technology; exploiting intrinsic attributes in molecular level, such as photochromism,18) could be one interesting resource. Meanwhile, chaotically oscillating dynamics have been utilized in recent intelligent functions such as decision making,12) Monte-Carlo computation,19) and artificial data generation through generative adversarial networks;20) these applications indicate the possibility of directly utilizing random physical processes in nature.

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