INTRODUCTION

Single-photon sources (SPSs) that can operate at room temperature and telecommunication wavelengths are promising candidates for on-chip quantum light sources for quantum computing and quantum cryptography because a single photon can be generated at both room temperature and telecommunication wavelengths on silicon chips. However, for the applications of quantum information, such as quantum computing and quantum cryptography, higher performance SPSs that exhibit both high purity and high efficiency of single-photon generation are required. Here, we theoretically propose high-performance SPSs that simultaneously achieve high-purity and high-efficiency single-photon generation by using short and functionalized air-suspended SWCNTs. The simulated exciton dynamics, time-resolved photoluminescence, and photon correlation properties indicate that exciton—exciton annihilation, end quenching, and trapping in the defect introduced by functionalization such as oxygen or aryl doping play important roles in determining the emission and single-photon properties, which strongly depend on SWCNT length and excitation intensity. We found that high performance SPSs that exhibit simultaneously high single-photon purity of 99.87% and high single-photon generation efficiency of 99.84% can be realized by using air-suspended functionalized SWCNTs with a length of approximately 100 nm under high excitation conditions. This ideal SPS can enable high rate and long-distance quantum key distributions at room temperature.

KEYWORDS: single-walled carbon nanotubes, single-photon sources, aryl-sp³ defect, photoluminescence, exciton localization

ABSTRACT: A single-photon source (SPS) based on a single-walled carbon nanotube (SWCNT) is a promising candidate for uncooled on-chip quantum information optoelectronics because a single photon can be generated at both room temperature and telecommunication wavelengths on silicon chips. However, for the applications of quantum information, such as quantum computing and quantum cryptography, higher performance SPSs that exhibit both high purity and high efficiency of single-photon generation are required. Here, we theoretically propose high-performance SPSs that simultaneously achieve high-purity and high-efficiency single-photon generation by using short and functionalized air-suspended SWCNTs. The simulated exciton dynamics, time-resolved photoluminescence, and photon correlation properties indicate that exciton—exciton annihilation, end quenching, and trapping in the defect introduced by functionalization such as oxygen or aryl doping play important roles in determining the emission and single-photon properties, which strongly depend on SWCNT length and excitation intensity. We found that high performance SPSs that exhibit simultaneously high single-photon purity of 99.87% and high single-photon generation efficiency of 99.84% can be realized by using air-suspended functionalized SWCNTs with a length of approximately 100 nm under high excitation conditions. This ideal SPS can enable high rate and long-distance quantum key distributions at room temperature.

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INTRODUCTION

Single-photon sources (SPSs) that can operate at room temperature and telecommunication wavelengths are promising candidates for on-chip quantum light sources for quantum computing and quantum cryptography because a single photon can be generated at both room temperature and telecommunication wavelengths on silicon chips. However, SPSs based on solid-state semiconductors face major operation challenges at both room temperature and telecommunication wavelengths. Single-photon sources (SPSs) based on solid-state semiconductor quantum dots on silicon-based platforms. Quantum key distribution for long distances has been reported by using air-suspended pristine SWCNTs.3 Theoretical studies of SPS from air-suspended pristine SWCNTs show that long exciton diffusion length due to the clean environment of the air-suspended SWCNTs and exciton—exciton annihilation (EEA) play important roles in SPE.8 Pristine air-suspended SWCNT's exhibit a higher quantum efficiency of photoluminescence (PL) than suspended SWCNTs on substrates because they do not experience environmental effects due to substrates and surfactants.11-20 However, it is very difficult for pristine SWCNTs to achieve a high purity SPE of 80% or more (i.e., g²(0) < 0.2) owing to weak localization of excitons, as reported experimentally and theoretically.10 Various efforts have been made to develop methods for SPE from SWCNTs,1,2,21-27 and high single-photon purity was recently achieved by introducing an aryl sp³ defect into dispersion SWCNTs1,28-30. In these reports, high purity SPE can be obtained by the localization of a single exciton at the defect. This is a very attractive approach because the extremely pure SPE with a small value of the second-order photon correlation functions at zero delay time g²(0) ~ 0.01 can be obtained from the unsuspended SWCNTs on substrates. However, as the single-photon generation efficiency was at...
most 13%, it may be difficult to simultaneously achieve both high single-photon purity and single-photon generation efficiency by simply introducing a defect site in the SWCNTs. At low temperatures, studies using resonators and pristine SWCNTs have reported that the single-photon generation efficiency of 62% and 40% has been achieved, and $g(0) = 0.3$ and 0.03,26,27 Hence, a novel method that can realize both high purity and high efficiency of SPE at room temperature should be developed for applications of quantum information such as quantum key distribution.

In this paper, we theoretically present SPSs that exhibit both high-purity and high-efficiency single-photon generation by shortening and functionalization of air-suspended SWCNTs. We simulated the time-resolved exciton dynamics and PL and photon correlation for pristine and functionalized air-suspended SWCNTs as parameters of SWCNT length and excitation intensity to investigate the exciton dynamics and their effect on SPS performance. The simulated results of exciton dynamics and photon correlation properties indicate that EEA, end quenching, and trapping of one exciton in a defect play important roles in influencing the PL decay and SPE properties, and these properties strongly depend on the SWCNT length and excitation intensity. We found that high performance SPSs with both high single-photon purity of 99.87% and high single-photon generation efficiency of 99.84% can be realized by using short (100 nm) air-suspended functionalized SWCNTs under high excitation conditions.

## RESULTS AND DISCUSSION

In this study, we investigated the dynamics of excitons generated by photoexcitation in one-dimensional air-suspended SWCNTs based on a Monte Carlo simulation as reported previously.10,31–33 Briefly, the air-suspended carbon nanotube was excited with a pulsed laser, which was focused on the center of the SWCNTs with a spot diameter of 500 nm. The generated excitons were stochastically distributed according to the Gaussian profile of the excitation laser. The number of generated excitons obey the Poisson distribution

$$P_{k}(n_g) = \frac{N_g^{n_g}}{n_g!} \exp(-N_g)$$

(1)

where $n_g$ is the number of generated excitons and $N_g$ is the average $\langle n_g \rangle$. After the generation of excitons, the exciton diffusion was calculated using the Monte Carlo method. The diffusion length $l$ was taken as 300, 500, and 1000 nm, which are diffusion lengths of suspended carbon nanotubes.15,34 The SWCNT length $L$ varied from 50 to 4000 nm. At both ends of the SWCNT, nonradiative relaxation occurred by end quenching. When two excitons meet, EEA occurs, and one exciton disappears (green arrows). (b) Schematic of the three-level model in pristine SWCNTs. IB, ID, and IT mean bright, dark, and final states, respectively. Bright and dark excitons recombine at the rates of $k_B$ and $k_D$, respectively. Thermalization between dark and bright states can occur at rates $k_0$ and $k_{DT}$. The free exciton is in a trapped state when the exciton moves to the defect site, and the IT state is unoccupied ($k_{DT}$ and $k_{BT}$). The trapped excitons recombine at a rate of $k_T$.

![Figure 1](image)

**Figure 1.** (a) Schematic view of the dynamics of excitons on the aryl-functionalized SWCNT. When a free exciton moves to the defect site, the exciton is trapped into the defect (blue arrow) and can no longer be detrapped. At both ends of the SWCNT, nonradiative relaxation occurs by end quenching (red arrow). When two excitons meet, EEA occurs, and one exciton disappears (green arrows). (b) Schematic of the three-level model in pristine SWCNTs. IB, ID, and IT mean bright, dark, and final states, respectively. Bright and dark excitons recombine at the rates of $k_B$ and $k_D$, respectively. Thermalization between dark and bright states can occur at rates $k_0$ and $k_{DT}$. The free exciton is in a trapped IT state when the exciton moves to the defect site, and the IT state is unoccupied ($k_{DT}$ and $k_{BT}$). The trapped excitons recombine at a rate of $k_T$. We assumed that each functionalized SWCNT has one defect. For a three-level system, the ground (final) state |F⟩, bright |B⟩, and dark |D⟩ exciton states are considered, and the trap state |T⟩ is additionally considered for a four-level system. We assume that the relaxation times of bright and dark excitons are $1/k_B = 50$ ps and $1/k_D = 500$ ps, respectively.31 We neglected radiative recombination from the dark state.16 Thermalization between dark and bright states can occur, and the transition rates are given by $k_{up} = g_D n$, $k_{down} = g_B (n + 1)$, where $g_D$ is the zero-temperature bright to dark transition rate, and $n$ is the density of dark excitons. In the defect, an exciton is trapped by the |T⟩ state, whose energy is lower than the free exciton levels by about 10 meV, which is enough to prohibit the exciton from returning to higher levels.26 This trapped exciton has a relaxation time of $1/k_T = 300$ ps as previously reported for aryl-functionalized or oxygen-doped SWCNTs.1,2,29 As the trapped exciton is spatially localized with a length scale of 1–3 nm,2,29 the trap state is treated as a point defect without the spatial distribution in our simulation. The dark exciton level is ignored in the simulation. The dark exciton level is ignored in the simulation. The dark exciton level is ignored in the simulation.
higher than the bright one, and the relaxation rate from the dark states is much slower than that from the bright state in the defect as reported for functionalized SWCNTs by aryl sp³ defects.29,39 Using the above methods, we can calculate exciton relaxations, i.e., the radiative relaxations from the bright state and the trap state, and the nonradiative relaxations from the dark state, EEA, and the end quenching. We note that these calculations aid in understanding exciton dynamics and could lead to improvement of emission properties because it is very difficult to experimentally distinguish the time-resolved results of these different nonradiative relaxations.

Figure 2a shows the calculated results of radiative recombination from bright excitons and nonradiative recombinations from dark excitons (black), EEA (green), and the end quenching (blue) for pristine SWCNTs with an average number of initially generated excitons: \( N_g = 25 \), SWSNT length \( L = 3 \) \( \mu \)m, and diffusion length of excitons \( l = 500 \) nm. The vertical axis indicates the number of events that occurred per 0.1 ps. (b) The \( L \) dependence simulation result of PL decay for pristine SWCNTs with \( N_g = 25 \) and \( I = 500 \) nm. \( L = 1 \) \( \mu \)m (green), 2 \( \mu \)m (purple), 3 \( \mu \)m (red), and 4 \( \mu \)m (blue) are used. The curves are fitted with double or triple exponential functions (black). (c) The \( N_g \) dependence of PL decay for pristine SWCNTs with \( L = 3 \) \( \mu \)m nm and \( I = 500 \) nm. \( N_g = 100 \) (green), 25 (red), and 0.5 (blue) are used. (d) The \( l \) dependence of PL decay for pristine SWCNTs with \( L = 3 \) \( \mu \)m and \( N_g = 25 \). \( l = 300 \) nm (blue), 500 nm (red), and 1000 nm (green) are used. These curves are fitted with double or triple exponential functions (black).
decay curves can typically be fitted by a triple-exponential function \( f(t) = A_{\text{EEA}} \exp(-t/\tau_{\text{EEA}}) + A_{\text{Int}} \exp(-t/\tau_{\text{Int}}) + A_{\text{EQ}} \exp(-t/\tau_{\text{EQ}}) \), where \( A_{\text{EEA}}, A_{\text{Int}} \) and \( A_{\text{EQ}} \) are coefficients (decay time) in the regions of EEA, intermediate, and end quenching (shown as \( R_{\text{EEA}}, R_{\text{Int}} \) and \( R_{\text{EQ}} \) in Figure 2b), respectively. Immediately after excitation (i.e., in the initial \( R_{\text{EEA}} \) region in Figure 2b), these curves exhibit a fast decay with the decay time of \( \sim 4 \) ps, which corresponds to the EEAs relaxation as shown by the green curve in Figure 2a. This fast EEAs relaxation time of a few picoseconds is consistent with the results of previous papers.\(^{40,41}\) The decay curve in the \( R_{\text{EEA}} \) region cannot be fitted using a single-exponential function because the EEAs rate strongly depends on the exciton density as mentioned above.

Following \( R_{\text{EEA}} \), an intermediate region \( R_{\text{Int}} \) which arises from the relaxations of bright excitons and end quenching, and a subsequent region \( R_{\text{EQ}} \), which is due to the end quenching, appear as shown in Figure 2b. End quenching plays an important role in the PL decay in the \( R_{\text{Int}} \) and \( R_{\text{EQ}} \) regions, where time-resolved recombination by end quenching has large counts as shown in Figure 2a. For several-\( \mu \)m-length SWCNTs, the rate of recombination due to end quenching is faster than the rate of recombination from the dark state. As a result, the population of dark excitons, from which bright excitons are excited, is mainly dominated by end quenching, and the PL decay in the \( R_{\text{EQ}} \) region is mostly determined by end quenching rates. In fact, the PL decay curve for a 3 \( \mu \)m length SWCNT, shown by the red curve in Figure 2b, has a decay time of 195 ps, which corresponds to the decay time of the end quenching obtained from the blue curve in Figure 2a. The intermediate region \( R_{\text{Int}} \) which has a decay time of several tens of picoseconds, is related to both end quenching and the radiative recombination of bright excitons, which is shown as \( k_{\text{B}} \) in Figure 1b. Assuming that the PL decay time in the \( R_{\text{EQ}} \) region can be calculated by the superposition of the \( R_{\text{EEA}} \) decay rate \( (1/195 \text{ ps}^{-1}) \) and \( k_{\text{B}} \) \((1/50 \text{ ps}^{-1} \text{ in this simulation})\), this PL decay time is estimated to be 40 ps, which corresponds to the PL decay time obtained from the fitting of the red curve in the \( R_{\text{Int}} \) region of Figure 2b.

As shown in Figure 2b, the decay curve changes depending on the SWCNT length; i.e., the short SWCNT \((L = 1 \text{ \mu m})\) exhibits a double-exponential decay curve, although the long SWCNT exhibits a triple-exponential decay curve as mentioned above. This is because \( R_{\text{EQ}} \) is not distinguishable from \( R_{\text{Int}} \) when the relaxation time of end quenching for a short SWCNT becomes shorter than the relaxation time of a bright exciton of \( 1/k_{\text{B}} \approx 50 \text{ ps} \). In fact, the decay time of \( R_{\text{EQ}} \) is shortened with decreasing SWCNT length as shown in Figure 2b \((268, 195, 119, \text{ and } \approx 23 \text{ ps for } L = 4, 3, 2, \text{ and } 1 \text{ \mu m}, \text{ respectively})\).

Figure 2c shows the excitation intensity \( N_e \) dependence of PL decay curves for pristine SWCNTs at \( L = 3000 \text{ nm} \) and \( L = 500 \text{ nm} \). In the \( R_{\text{Int}} \) and \( R_{\text{EQ}} \) regions, there is no significant difference in the decay curves at low and high excitation intensities \((N_e = 0.5 \text{ and } 100, \text{ respectively})\). However, in the \( R_{\text{EEA}} \) region, the decay behavior of EEA disappears at low excitation intensities (see inset), and the decay curve at low excitation intensities can be fitted using a double-exponential function because a small amount of EEA occurs at lower exciton densities.\(^{39,40,42-45}\) Figure 2d shows the diffusion length \( L \) dependence of PL decay curves at \( N_e = 25 \) and \( L = 3000 \text{ nm} \). The decay time in the \( R_{\text{EEA}} \) region is shortened with increasing \( L \) because the excitons can reach the ends of the SWCNTs more quickly at longer \( L \). The decay times at diffusion lengths of 300, 500, and 1000 nm are 328, 195, and 75 ps, respectively.

In this study, we theoretically investigated the properties of single-photon generation of a pristine SWCNT, especially the purity and efficiency of single-photon generation. Briefly, to simulate the photon correlation measurement and the efficiency of single-photon generation, we calculated the second-order correlation function at zero delay \( g^x(0) \), which takes zero for ideal SPE, and the average number of generated photons per pulse \( \langle n \rangle \). \( g^x(0) \) is given by\(^{10}\)

\[
g^x(0) = \frac{V - N}{N^2} + 1
\]

where \( N \) and \( V \) are the average and the dispersion of the number of photons generated for each excitation, respectively.

**Figures 3a and 3b** show the SWCNT length \( L \) dependence of \( g^x(0) \) and \( \langle n \rangle \) for pristine SWCNTs with \( L = 1000 \text{ nm} \) at low

\[ \langle n \rangle = 0.5 \text{ and high } N_e = 100 \] excitation intensities. A smaller \( g^x(0) \) indicates a higher single-photon purity because \( 1 - g^x(0) \) represents the single-photon purity. As shown in Figure 3a, the \( g^x(0) \) value becomes high at high excitation intensities because the annihilation of excitons by EEA is not sufficient, and multiple photons are generated from the residual excitons, which are free excitons remaining in the SWCNT after the first photon emission. Under the weak excitation condition, shorter SWCNTs tend to have a higher \( g^x(0) \) and lower \( \langle n \rangle \) than longer SWCNTs because end quenching is more likely to occur in shorter SWCNTs. For the single-photon generation from the SWCNT, EEA plays an important role in generating a single exciton and a quantum light such as a single photon; therefore, excessive nonradiative recombination by end quenching worsens the purity and efficiency of SPE. As shown in Figure 3b, \( \langle n \rangle \) decreases with decreasing \( L \) because of an increase in the end quenching under the strong and weak excitation conditions. The results shown in Figure 3a indicate that the minimum \( g^x(0) \) for the pristine SWCNTs is \( \sim 0.4 \), which is consistent with previously reported experimental and theoretical \( g^x(0) \), and this implies that high purity and efficiency SPEs may not be achieved by using the pristine SWCNTs.
To realize SPSs with higher purity and efficiency, we theoretically investigated the SPE from air-suspended functionalized SWCNTs. In the functionalized SWCNTs, the defect creates a deep trap state below the original bright state. The energy of the trap state is sufficiently lower than the original bright state; this indicates that the light emission from the trapped exciton in the defect can be separated by using optical band-pass filters. Therefore, in the calculation for functionalized SWCNTs, \( N \) and \( V \) can be calculated only for photons emitted from the trapped excitons. Excitons are locally confined to the defect, which is formed at the center of a SWCNT in our simulation, as shown in Figure 1a, and this confined single exciton recombines and subsequently emits a single photon.

Figure 3c shows the \( L \) dependences of \( g^2(0) \) for functionalized SWCNTs. The defect site can trap only one exciton and two or more excitons cannot be simultaneously trapped owing to state filling, the probability of multiphoton generation is suppressed. Hence, the \( g^2(0) \) value for functionalized SWCNTs is drastically improved at all lengths and excitation intensities compared to that of the pristine SWCNTs. From the length dependence of \( g^2(0) \) for functionalized SWCNTs, it can be seen that the short functionalized SWCNTs have an extremely low \( g^2(0) \) under strong and weak excitation conditions.

Figures 4a and 4b give a schematic illustration of the photon generation mechanism in a functionalized SWCNT and explain how excitons diffuse and radiatively or nonradiatively recombine. In long functionalized SWCNTs (Figure 4a), excitons diffuse after exciton generation (L1), and end quenching, EEA, and defect trapping occur (L2). After the first photon emission from the trapped exciton in the defect (L3), a residual free exciton can be trapped again into the defect and can emit a second photon (L4–L6), causing an increase in \( g^2(0) \). As residual free excitons can easily remain owing to little end quenching, the long functionalized SWCNTs have a high \( g^2(0) \). On the other hand, in the short functionalized SWCNTs (Figure 4b), excitons are trapped in the defect or immediately recombine nonradiatively by end quenching (S1–S2). As it is difficult for excitons to survive owing to end quenching, the second photon emission is effectively suppressed, therefore causing an extremely low \( g^2(0) \).

Figure 3c also show the excitation intensity \( N_e \) dependence of \( g^2(0) \) for functionalized SWCNTs. For long functionalized SWCNTs, many excitons are generated under strong excitation conditions, and these free excitons can be trapped again, causing an increase in \( g^2(0) \) with increasing high excitation intensity (Figure 4a). However, for short functionalized SWCNTs, such a tendency is suppressed because the end quenching can efficiently eliminate excess free excitons (Figure 4b). Hence, short functionalized SWCNTs can maintain low \( g^2(0) \) values under strong excitation conditions; e.g., functionalized SWCNTs with \( L = 100 \) nm have \( g^2(0) = 0.00128 \pm 0.00001 \) under the strong excitation condition of \( N_e = 100 \). The \( L \) and \( N_e \) dependences of \( n_e \) for air-suspended functionalized SWCNTs are shown in Figure 3d. Interestingly, under strong excitation conditions, the functionalized SWCNT can maintain high values of \( n_e \) \( \sim 1 \) in the short length range owing to efficient single-photon generation and end quenching, in contrast to pristine SWCNTs, as shown in Figure 3b. This indicates great advantages that the air-suspended functionalized short SWCNTs (\( L = 100 \) nm) exhibit both high purity (low \( g^2(0) \sim 0.00128 \pm 0.00001 \)) and high efficiency (high \( n_e \) \( \sim 0.99842 \pm 0.00001 \)) under high excitation conditions, corresponding to an ideal SPS (as shown by the green arrows in Figures 3c and 3d).

Figure 5a shows the \( N_e \) dependence of the \( g^2(0) \) for short (100 nm) and long (4000 nm) functionalized SWCNTs at \( L = 1000 \) nm. The short SWCNTs (red circles) take a single-photon purity \( g^2(0) \) about two orders smaller than that of long SWCNTs (black circles). In particular, it should be noted that high single-photon purity \( g^2(0) \sim 0.001 \) can be maintained even at a high excitation intensity in the short air-suspended functionalized SWCNTs. Figure 5b shows the \( N_e \) dependence of the \( n_e \) for short and long functionalized SWCNTs. In the case of short functionalized SWCNTs, \( n_e \) is low under weak excitation intensity. However, when the excitation intensity increases, the average number of photons approaches almost one \( (n_e = 0.99842 \pm 0.00001) \) at \( N_e = 100 \). These results indicate that the short air-suspended functionalized SWCNTs can realize both a high single-photon purity of 99.87% \( g^2(0) = 0.00128 \pm 0.00001 \) and single-photon generation efficiency of 99.84% under strong excitation conditions. The SPS with an air-suspended short functionalized SWCNT is ideal for the quantum key distribution because ideal SPSs with high purity and efficiency enable long-distance and high-speed communication close to the theoretical limit.

The exciton dynamics in functionalized SWCNTs shown in Figure 4 is supported by the calculated time-resolved PL from the defect state shown in Figure 6. As an exciton trapped in the
defect is not eliminated by end quenching or EEA, the decay curves can be fitted using a single-exponential function rather than the triple-exponential function for pristine SWCNTs. The relaxation time was 293 ps for long and short functionalized SWCNTs. As the relaxation time from the defect level $1/k_T$ was set to 300 ps in this simulation, the decay time of PL was dominated by the relaxation time of the defect level.

It should be noted that the initial decay curves are different between short and long functionalized SWCNTs as shown in Figure 6. For the short functionalized SWCNTs, the initial PL intensity quickly increases within 0.1 ps and subsequently decreases monotonically. On the other hand, for the long functionalized SWCNTs, the initial PL intensity gradually increases for 20 ps. This difference in the initial PL can be attributed to the existence of residual free excitons, which can become trapped again into the defect and repetitively generate photons after recombination of the first trapped exciton (see Figure 4a L4–L6). Although most free excitons are quickly eliminated by end quenching in a short functionalized SWCNT, many residual free excitons exist in a long functionalized SWCNT because long diffusion is necessary for end quenching. This can also be found in the time-resolved count of nonradiative recombination by end quenching as shown in Figure 7. For long functionalized SWCNTs, the end-quenching count initially increases for about 40 ps and subsequently decays with a time constant of about 70 ps. This delay of the decay curve is caused by the long diffusions in the long SWCNT. On the other hand, for short functionalized SWCNTs, end quenching effectively occurs immediately after the excitation of excitons within 2 ps because of its short...
diffusion. This indicates that the free excitons, which were not trapped in the defect, immediately recombine nonradiatively by end quenching in the short SWCNTs as shown in Figure 4b. This exciton dynamics of short functionalized SWCNTs can enable realization of high performance SPSs with both high purity and high efficiency of single-photon generation as shown in Figure 3 and Figure 5.

Figure 8 shows the \( L \) and \( N_g \) dependence of \( \langle n \rangle \) and \( g^2(0) \) for functionalized SWCNTs with \( l = 1000 \) nm, and we list the

![Figure 8](https://example.com/figure8.png)

**Table 1.** \( L \) and \( N_g \) Dependence of \( g^2(0) \) and \( \langle n \rangle \) for \( l = 1000 \) nm

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<th>( L ) (nm)</th>
<th>( N_g )</th>
<th>( \langle n \rangle )</th>
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typical values of our simulation in Table 1. Importantly, the optimal length of the functionalized SWCNT for a high performance SPS, which should enable both high-purity and high-efficiency single-photon generation, depends on the excitation intensity \( N_g \). For example, for \( N_g = 100 \) and 25, the optimal lengths of the SWNTs are 100 and 400 nm, respectively, if the optimal length is determined by the condition that \( \langle n \rangle \) is more than 0.99 and \( g^2(0) \) is as small as possible. As a shorter functionalized SWCNT has a lower \( \langle n \rangle \) value owing to nonradiative recombination by end quenching, the efficiency of single-photon generation will be less than 99% when the length of the SWCNT is shorter than the optimal length. On the other hand, longer functionalized SWCNTs have higher \( g^2(0) \) values. These findings indicate that ideal SPSs cannot be achieved with too long or too short SWCNT lengths. As shown in Figure 8, the optimal length for the strong excitation condition is shorter than that for the weak excitation condition. This is because \( \langle n \rangle \) can be maintained at \( \sim 1 \), which indicates that one exciton can be certainly trapped under one pulse of excitation light, for short SWCNTs under strong excitation conditions, and \( g^2(0) \) can decrease with decreasing SWCNT length. Under our excitation intensities (\( N_g = 0.5 \)– 100), the longest air-suspended functionalized SWCNT length is 400 nm, satisfying \( g^2(0) \sim 0.01 \) and \( \langle n \rangle \sim 0.99 \). At this time, the excitation intensity was \( N_g = 25 \) (see Table 1).

**CONCLUSIONS**

We theoretically investigated SPSs with high purity and high efficiency of single-photon generation by using short air-suspended functionalized SWCNTs. The 100 nm length air-suspended functionalized SWCNTs can achieve SPSs with a high single-photon purity of 99.87% and high single-photon generation efficiency of 99.84%. As SPSs with SWCNTs can work at room temperature and the telecommunication wavelength on silicon chips, ideal SPSs, which can realize on-chip, room-temperature quantum devices, can be fabricated. Furthermore, the SWCNT-based high-performance SPSs can enable a high-rate and long-distance quantum key distribution based on the integrated SPSs at room temperature.

**METHODS**

**Calculation of Exciton Dynamics, Time-Resolved PL, and Photon Correlation.** We calculated the exciton dynamics and PL in SWCNTs after photoexcitation by Monte Carlo simulations, taking into account the exciton relaxations shown in Figure 1. The time resolution is 0.1 ps for all time-resolved measurements except those shown in Figure S1, and the calculation is ended when the emission photon count reaches \( 10^6 \). As the time-resolved measurements of nonradiative recombination are difficult to obtain in experiments, these results help us to understand how dark excitons, EEA, and end quenching affect the radiative recombination in the bright state. As the trap state is sufficiently and energetically lower than the original bright state, the emission from the trap state can be experimentally separated from that from free excitons by using a band-pass filter or monochromator. Hence, photon correlation is calculated only for photons from the trapped excitons.

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsanm.9b02209.

Experimental and simulated PL decay curves of pristine SWCNTs, list of constants used for our simulation, and 2D phase diagram of \( \langle n \rangle \) and \( g^2(0) \) with various ratios of diffusion length \( l \) to pristine and functionalized SWCNT length \( L \). (PDF)

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Notes
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