Ultraviolet phosphorescent carbon nanodots

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Abstract
Phosphorescent carbon nanodots (CNDs) have generated enormous interest recently, and the CND phosphorescence is usually located in the visible region, while ultraviolet (UV) phosphorescent CNDs have not been reported thus far. Herein, the UV phosphorescence of CNDs was achieved by decreasing conjugation size and in-situ spatial confinement in a NaCNO crystal. The electron transition from the px to the sp² orbit of the N atoms within the CNDs can generate one-unit orbital angular momentum, providing a driving force for the triplet excitons population of the CNDs. The confinement caused by the NaCNO crystal reduces the energy dissipation paths of the generated triplet excitons. By further tailoring the size of the CNDs, the phosphorescence wavelength can be tuned to 348 nm, and the room temperature lifetime of the CNDs can reach 15.8 ms. As a demonstration, the UV phosphorescent CNDs were used for inactivating gram-negative and gram-positive bacteria through the emission of their high-energy photons over a long duration, and the resulting antibacterial efficiency reached over 99.9%. This work provides a rational design strategy for UV phosphorescent CNDs and demonstrates their novel antibacterial applications.

Introduction
Carbon nanodots (CNDs) as promising phosphorescence candidates develop rapidly.¹,² Their relatively easy preparation process, unique optical properties, and high biocompatibility make them especially fascinating in the field of bioscience and technology.³⁻⁵ To date, room temperature phosphorescent CNDs have facilitated amazing advances in optoelectronics devices, time-resolved imaging, and phosphorescence bioimaging, due to the efforts of scientists.⁶⁻¹¹ However, most phosphorescence of CNDs is centered in the visible region, and there remains an issue of limited phosphorescence wavelengths. In particular, ultraviolet (UV) phosphorescent CNDs that can emit high-energy photons over a long duration have not been realized. Thus, the development of UV phosphorescent CNDs is of great importance and significance considering their attractive applications in UV light-emitting devices, sterilization and so on.¹²⁻¹⁴

Modulating the wavelength and brightening the triplet excitons of CNDs are the key points for inducing UV phosphorescence. The conjugation size modulation of CNDs as the most accepted method has been used to produce colorful, glowing CNDs. Enlarging the conjugation size of CNDs can lead to longer wavelength emissions due to the decrease in system energy caused by the increase in electron delocalization. The corresponding results have been reported by Qu and Lin et al., and this is significant for near-infrared emissive CNDs.¹⁵,¹⁶ Crosslink-enhanced emission (CEE), a novel concept conceived by Yang and coauthors, sheds light on the emission mechanism of CNDs.¹⁷ Cross-linking during the formation of CNDs can also increase their conjugate size to some extent, leading to a narrower bandgap of the synthesized CNDs. Thus, optimizing the carbonization process and decreasing the conjugation size of CNDs will endow them with the desired UV emission properties. As for brightening triplet excitons, “aggregation”, “crystallization” and “confined-domain CEE” concepts were proposed for aggregation-induced emission materials, molecule emission materials, and CND emission materials, the study of which is important for realizing their phosphorescence and understanding the underlying...
mechanisms. The nature of the three concepts is similar, that is, the restriction of the vibration/rotation of the luminophore, which can be described as confinement. The confinement of CNDs within nano spaces has been used for water-soluble phosphorescent CNDs. Thus, UV phosphorescent CNDs can be achieved by combining confinement and conjugation size modulation engineering.

We aim to achieve the UV phosphorescence of CNDs in this work; thus, the formed CNDs should have a small conjugation size and sufficient ISC driving force within a confined environment. According to calculations of the first-principles density functional theory, the bandgaps of configurations consisting of melamine molecules are in the region of 3.7–3.8 eV, indicating that CNDs with a small conjugation size are expected to be formed by using melamine as a precursor. In addition, N atoms with lone pairs of electrons favor the triplet exciton population. By tailoring the size of the CNDs from over 100 to ~3.6 nm, the conjugation size decreases, the phosphorescence emission peak can be tuned to 348 nm, and the lifetime can reach 15.8 ms under the condition of NaCNO. Due to their high-energy photons and long-lasting time, the CNDs are used to inactivate gram-negative/positive bacteria with antibacterial efficiency exceeding 99.9%, demonstrating their novel antibacterial applications.

Results

UV phosphorescent CNDs were prepared by using melamine as the luminophore in terms of electron orbits, which will be discussed carefully below. As indicated in Fig. 1a, the outer-shell electron distributions of the carbon and nitrogen atom orbits are 2s2p2 and 2s2p3, and one s orbital and two p orbitals form three sp2 hybrid orbitals in the CNDs. For the nitrogen atoms within the CNDs, the three sp2 hybrid orbitals are not equivalent, which leads to a lone pair of electrons residing in one of the sp2 hybrid orbitals. This lone pair of electrons is not involved in the n-electron system, thus strengthening the spin-orbit coupling (SOC) because of the inversion asymmetry band structure, and this is beneficial for the realization of phosphorescence. Generally, SOC plays a dominant role in electron spin-flipping in heavy-atom free systems. From the El-Sayed rule, the process of the ISC transition of the \( 3\text{(n, } \pi^* \text{)} \rightarrow \text{3}\text{(n, } \pi^* \text{)} \) and \( 3\text{(n, } \pi^* \text{)} \rightarrow \text{3}\text{(n, } \pi^* \text{)} \) is easy to implement. However, the ISC of the \( 3\text{(n, } \pi^* \text{)} \rightarrow \text{3}\text{(n, } \pi^* \text{)} \) with similar electronic configurations is not effective due to the spin-forbidden transition, which leads to the low efficiency of SOC. Therefore, for the recombination rate of triplet excitons, the \( 3\text{(n, } \pi^* \text{)} \) triplet has a slow recombination rate compared to \( 3\text{(n, } \pi^* \text{)} \) due to the spin-forbidden transition. In this system, the n orbit is one of the sp2 orbitals of the N atom, and the electron transition from the sp2 to the px orbit or the px to the sp2 orbit of the N atom will produce a one-unit change in the orbital angular momentum, meeting the required angular momentum of the electron spin-flip (from −1/2 to 1/2; or 1/2 to −1/2), which is necessary for the triplet excitation population of the CNDs. The interaction between the orbit and the spin can be described by the equations in Fig. 1a. The spin-orbit coupling Hamiltonian operator, which is in direct proportion to the product of the magnetic moment of orbit angular momentum \( \mu_\text{L} \) and spin angular momentum \( \mu_\text{S} \). The spin-orbit coupling energy \( E_\text{soc} \) pertains to the overlap of the initial orbit wave functions \( \psi_1 \) and terminative orbit wave functions \( \psi_2 \). A larger \( E_\text{soc} \) indicates stronger spin-orbit coupling. A change in spin angular momentum \( \mu_\text{S} \) induces orbit distortion, and a change in orbit angular momentum \( \mu_\text{L} \) can also lead to electron spin-flipping. Thus, the distinguishing feature of \( H_\text{soc} \) is that it can rotate the corresponding electron wave functions by 90°. In this system, matrix-\( (px|H_\text{soc}|sp^2) \), has a large value under the effect of \( H_\text{soc} \). Furthermore, the bandgaps of CNDs with different structures were investigated based on first-principles density functional theory calculations. Possible configurations denoted as configuration-1 and configuration-2, are shown in Fig. S1. The bandgaps of the two configurations are 3.769 and 3.801 eV. The high overlap of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) leads to the effective recombination of electrons and holes, endowing the CNDs with efficient UV luminescence. To realize UV phosphorescence, the CNDs were placed into a rigid environment provided by the NaCNO crystal to stabilize the produced triplet excitons. The plot of the phosphorescence spectra versus time illustrated in Fig. 1b shows the phosphorescence peaks at ~348 nm, which is the first report of UV phosphorescent CNDs. The lifetime of the CNDs is ~15.8 ms. The total photoluminescence (PL) quantum yield (QY) is about 31.7% and the phosphorescence QY is about 16.2%.

The corresponding morphology and structure evolution of the CNDs were characterized by transmission electron microscopy (TEM), as shown in Fig. 2a–c. Large-scale TEM images indicate that the CNDs have different sizes (Fig. S2), which can clearly reveal the size evolution of the CNDs under different temperatures. Remarkably, the high-resolution TEM image in Fig. 2c shows a clear lattice spacing of 0.21 nm, indicating the formation of CNDs with high crystallinity. Fourier transform infrared (FTIR) spectra and X-ray diffraction (XRD) patterns were collected to further understand the surface and inner structures of the CNDs. A peak at ~27.4° can be observed from the XRD pattern, and the other peaks correspond to Na2CO3 (owing to the reaction between CO2 and NaOH) (Fig. S3a). With increasing temperature, some of the Na2CO3 may react with the amine groups in the
melamine to form NaCNO, the existence of which can be confirmed by the peak centered at 2207 cm$^{-1}$ in the FTIR spectra in Fig. S4. No obvious peaks of the CNDs were observed due to a lower proportion of the CNDs within the NaCNO (Fig. S3b, c). The NaCNO provided a rigid, confining, and isolating environment for the CNDs. An
Fig. 2 The formation process and fluorescence characterization. TEM images of crosslinked melamine at 200 °C (a), CNDs formed at 300 °C (b) and CNDs formed at 400 °C (c). The insets are the size distributions and high-resolution TEM images of the CNDs. d Schematic diagram of the formation mechanism of the CNDs under different conditions. Excitation-fluorescence mapping profiles of crosslinked melamine@Na₂CO₃ (e), CNDs-1@NaCNO (f), and CNDs-2@NaCNO (g). h The corresponding UV–vis, PL emission spectra of the crosslinked melamine@Na₂CO₃, CNDs-1@NaCNO, and CNDs-2@NaCNO.
-OH stretching vibrations signals located at 3374 cm⁻¹ can be observed, and the peak centered at 3451 cm⁻¹ could be assigned to the -NH₂ surface groups³⁶,³⁷. Additionally, the peak centered at ~1800 cm⁻¹ originates from the vibration of C=N⁰⁸. Obviously, from the XPS spectra shown in Fig. 5, the Na₂CO₃ was formed at 200 °C, Na₂CO₃ can react with amidogen, leading to the collision of the shell and the loss of nitrogen. Thus, a possible formation process of the UV phosphorescent CNDs based on these results is proposed in Fig. 2d. The melamine precursor underwent cross-linking through a polymerization reaction at 200 °C, Na₂CO₃ was formed around the crosslinked melamine due to the reaction between CO₂ and NaOH, and the size of the crosslinked melamine@Na₂CO₃ was over 100 nm. When the temperature increased to 300 °C, a highly dense amorphous carbon particle formed, and was accompanied by the formation of NaCNO, resulting in the production of CNDs@NaCNO. The size of the CNDs decreased to ~7.8 nm, and most of the CNDs in this step exhibited an amorphous structure. The high temperature will lead to the aromatization of amorphous carbon particles; when the synthesis temperature increased to 400 °C, the average size of the CNDs decreased to 3.6 nm.

Furthermore, the photophysical properties of the CNDs were investigated. The PL-excitation pattern shows that the luminescence peak at 430 nm changes little with the excitation wavelength, indicating that there is only one kind of luminescence center in the CNDs (Fig. 2e). For the CNDs prepared at 300 °C (denoted as CNDs-1@NaCNO), a luminescence peak was observed at ~360 nm (Fig. 2f). In the case of the CNDs prepared at 400 °C (denoted as CNDs-2@NaCNO), emission peaks were observed at ~342 nm (Fig. 2g). The total PL QYs of the samples synthesized at 200, 300, and 400 °C are 5.3, 12.6, and 31.7%, respectively. Notably, the CNDs showed obvious size-dependent luminescence, and the fluorescence peaks blueshifted from 430 to 342 nm. In detail, the blueshifting of the emission is due to the decreased electron delocalization size, resulting in the increased bandgap of the CNDs⁹⁰. In Fig. 2h, absorption and PL emission spectra show a clear blue shift with decreasing sample size. The absorption spectra demonstrate the n-π* transition from C=O and the π-π* transition from C=C in the range of 280 to 350 nm⁴⁰,⁴¹. The absorption peaks from the n-π* transition blueshift to 248 nm, and the π-π* transition blueshifts to 204 nm for CNDs-1@NaCNO, and the peaks further blueshift to 244 and 200 nm for CNDs-2@NaCNO.

It is important to explore the luminescence dynamic process of the CNDs in the UV region, therefore, the PL lifetime profiles of the CNDs@NaCNO synthesized at 300 and 400 °C were measured (Fig. S6). The PL lifetime profiles of CNDs-2@NaCNO indicate a lifetime of 6.4 ns, which is longer than that of CNDs-1@NaCNO (2.9 ns). A high crystallinity decreased the vibration and rotation of the bonding in the CNDs-2, which decreased the non-radiative dissipation of the carriers, thus increasing the PL QY of the CNDs. For a better understanding of the UV emission properties of CNDs-2@NaCNO, the corresponding synchronous scan spectra is shown in Fig. S7a. The maximum emission intensity at 342 nm corresponds to a strong UV emission caused by the 322 nm excitation. Similarly, at 312 nm with a step length of 30 nm, the highest emission intensity was observed at 342 nm (Fig. S7b). For completeness, the emission intensities variation of the corresponding synchronous scan spectra is shown in Fig. S7c; it is worth noting that the spectra show a dominant emission with a peak at approximately 342 nm. Thus, the CNDs synthesized at higher temperatures have smaller sizes, higher crystallinities and larger bandgaps, leading to decreases in vibration/rotation and exciton confinement, and thus, efficient UV emission is achieved.

Interestingly, the CNDs confined in the NaCNO matrix show UV phosphorescence in addition to UV fluorescence. Some -NH₂ on the CND surfaces were substituted by -OH with increasing synthesis temperature, and the hydroxyls can ionize to form multiple ionic bonds with Na⁺, as shown in Fig. 3a. To further study the phosphorescence properties, the time-resolved phosphorescence spectra of CNDs-1@NaCNO and CNDs-2@NaCNO were measured, as shown in Fig. 3b, c. The CNDs-1@NaCNO exhibited a luminescence peak at 417 nm with a lifetime of 61.1 µs under the excitation of 290 nm light (Fig. S8a), which originated from the recombination of triplet excitons. The triplet excited states of the CNDs can be efficiently stabilized through the strong confinement of the NaCNO crystal. The energy difference (ΔEST) between the fluorescence and phosphorescence of CNDs-1@NaCNO is 0.45 eV (Fig. S9a). For the CNDs-2 with a small size, the phosphorescence peak blueshifted to 348 nm, and the lifetime increased to 15.82 ms (Fig. S8b). The ΔEST of CNDs-2@NaCNO decreased to 0.12 eV (Fig. S9b), which is favorable for the triplet exciton population. The higher-crystallinity carbon core and small size of CNDs-2 further decreased the vibration/rotation and enlarged the bandgap of the CNDs, leading to efficient UV phosphorescence. For clarity, the emission lifetime decay spectra of crosslinked melamine@Na₂CO₃, CNDs-1@NaCNO, and CNDs-2@NaCNO collected at 430, 417, and 348 nm are shown in Fig. 3d. In addition to the blueshift of the emission, the lifetimes of the CNDs can be adjusted from nanoseconds to microseconds and milliseconds. In particular, the phosphorescence spectra in Fig. 3e revealed an excitation-independent phosphorescence property of
CNDs-2@NaCNO with the excitation wavelength ranging from 270 to 330 nm.

Lifetime is one of the most important parameters for phosphorescence; thus, the temperature-dependent phosphorescence lifetime decay curves at 348 nm of CNDs-2@NaCNO were recorded from 78 to 353 K to further study the influencing factor of the phosphorescence lifetime, as shown in Fig. 3f. The phosphorescence lifetimes (Fig. S10) increased gradually as the recording temperature decreased, showing a strong temperature-dependent phosphorescence property. The phosphorescence QY and lifetime are related to the rates of the corresponding radiative recombination and nonradiative recombination, which can be depicted by the following Equations:

\[
\tau_p = \frac{1}{[k_p + \Sigma k_n]} 
\]

(1)

\[
\Phi_p = \frac{k_p}{[k_p + \Sigma k_n]} 
\]

(2)

where \(\tau_p\) is the lifetime; and \(k_p\) and \(k_n\) are the radiative recombination and nonradiative recombination rates, respectively. The \(k_n\) of CNDs-2@NaCNO is 52.9 s\(^{-1}\) at room temperature and 4.88 s\(^{-1}\) at 78 K, as shown in
Salmonella conditions were determined by the analysis of CNDs-2@NaCNO. The long-lifetime UV photon cytotoxicity chosen to evaluate the antibacterial performance of CNDs-2@NaCNO was activated and emitted UV photons for a long time. Thus, the interaction between the CNDs-2@NaCNO and bacteria can cause the death of more bacteria. The sterilization efficiency can reach more than 90% within three cycles and it is close to 100% after five cycles, as shown in Fig. S14b, c. The above results indicate that in terms of long-lifetime UV photon production, the number of UV photons of CNDs-2@NaCNO gradually increased due to the number of triggers by increasing irradiation and CNDs-2@NaCNO can be used as an antibacterial nanomaterial. Furthermore, a relative antibacterial test with NaCNO as a control group was performed for S. aureus (Fig. S16), and the cytotoxicity effect of NaCNO on S. aureus was negligible. In addition, confocal laser scanning microscope (CLSM) images of the bacteria stained with propidium iodide (PI) and SYTO 9 fluorescent indicator are shown in Fig. 4d. The bacteria stained by the PI indicator with damaged membranes showed red emission, while bacteria with intact membranes showed green emission due to SYTO 9 staining. For the bacteria incubated with CNDs-2@NaCNO under excitation with pulsed light, most of them showed bright red fluorescence in comparison to other groups, demonstrating the high antibacterial ability of CNDs-2@NaCNO. Furthermore, the scanning electron microscopy images in Fig. 4e demonstrates the morphological changes of S. aureus and Salmonella. The bacteria treated with CNDs-2@NaCNO under excitation with pulsed light showed obvious membrane damage, while the bacteria in the control group remained intact structure. These results indicated that the inactivation of bacteria is due to the rupture of bacterial membranes caused by exposure to high-energy photons.

**Discussion**

In conclusion, we proposed design principles for UV phosphorescent CNDs: (1) The formed CNDs should have a small conjugation size, and first-principles density functional theory calculations can provide qualitative guidance for precursor selection; (2) Heteroatoms with lone pairs of electrons should be included in the CNDs to provide enough ISC driving force; and (3) An in-situ generated confinement environment is favorable considering the instability of triplet excitons. Of course,
Fig. 4 Demonstration of the UV phosphorescent CNDs for antibacterial applications. a Schematic diagram of the interaction between UV phosphorescence and bacteria. b Images of the flat colony counting results under different conditions. c Number of bacterial counts (CFU mL$^{-1}$) in panel (b). d CLSM images of S. aureus and Salmonella after treatment with CNDs-2@NaCNO under excitation of pulsed light for five cycles (one cycle: irradiation six times within 1 min, excitation light: 310 nm, 7 mW cm$^{-2}$, 0.1 Hz); scale bar is 20 µm. e SEM images of the S. aureus and Salmonella under different conditions; scale bar is 1 µm.
deeper physical effects should be further investigated. Under the guidance of the proposed design principles, UV phosphorescent CNDs with peaks centered at 348 nm and lifetimes of 15.8 ms have been realized. The phosphorescence wavelength can be tuned from 430 to 348 nm by decreasing the size of the CNDs and suppressing the vibration/rotation of the CNDs with the confinement of the triplet excitons by the NaCNO crystal. The highest UV phosphorescence QY of the CNDs reached 16.2%. As a demonstration, the UV phosphorescent CNDs were used to inactivate gram-negative/positive bacteria, and the antibacterial efficiency reached up to 99.9% against S. aureus and Salmonella. These results provide a rational design strategy for UV phosphorescent CNDs, and demonstrate promise for the potential antibacterial applications of CNDs.

Materials and methods

Chemicals and materials
Melamine and sodium hydroxide were purchased from Shanghai Macklin Chemistry Co. Ltd. and used without further purification.

Synthesis of crosslinked melamine@Na2CO3, CNDs-1@NaCNO, and CNDs-2@NaCNO
First, 0.5 g melamine and 0.1 g NaOH precursors were mixed in a mortar and ground uniformly for 10 min, and then the resulting solid mixtures of melamine and NaOH were calcined at 200 °C in a muffle furnace for 10 h with a heating rate of 20 °C min⁻¹. The generated products were denoted as crosslinked melamine@Na2CO3. For CNDs-1@NaCNO, the ratio between o-melamine and NaOH was 1:1 (0.5 g melamine and 0.5 g NaOH), and CNDs-1@NaCNO powders was obtained after heating at 300 °C for 10 h. For CNDs-2@NaCNO, the reaction temperature was further increased to 400 °C, and after 10 h, CNDs-2@NaCNO powders were obtained. The obtained sample powders were used for further experimentation.

First-principles density functional theory calculations
The optimization of the ground state geometries of the UV phosphorescent CNDs was performed through density functional theory (DFT), and identity approximation was carried out using the B3LYP functional together with the chain of spheres exchange method. Due to the different noncovalent interactions, Grimme’s dispersion correction was introduced in the calculation. Time-dependent density functional theory and Grimme’s dispersion (D3) correction were used to calculate the excitation energies. Further confirmation of the nature of the structure with minimum energy (resting point) was performed by conducting frequency analysis. DFT calculations were performed using the Visual Molecular Dynamics package and Gaussian 09 package.

Animal experiments
BALB/c mice (6–8 weeks, female) were purchased from Hunan Shrek Jingda experimental animal Co., Ltd. All the experiments were in compliance with the policies of the National Ministry of Health. All animal procedures were performed following a protocol approved by the Institute of Drug Discovery & Development of Zhengzhou University (syxk (yu) 2018-0004).

Histological experiments and cytotoxicity measurements
CNDs-2-NaCNO (1000 µg/ml) were injected intravenously into mice for 1 day and 7 days and the control group of mice was injected with normal saline. On the 7th day, the organs (heart, kidney, bladder, liver, and lung,) from all wound sites were excised for histopathological evaluation.

The cytotoxicity was measured via MTT cell proliferation-toxicity assays. Human umbilical vein endothelial (HUVEC) cells and human hepatocellular carcinoma (HepG2) cells were seeded onto 96-well plates at 37 °C. Twenty-four hours later, CNDs-2-NaCNO solutions of different concentrations (0–200 µg/mL) were added to each well. Subsequently, the MTT solution (10 µL) was added to each well of the plate after further incubation for 24 h. Then, the optical density properties at 492 nm of each well were recorded to evaluate the cell viability.

Antibacterial experiments
The antibacterial properties were evaluated by using S. aureus and Salmonella as gram-positive/negative bacteria. A single colony of S. aureus was added to a 20 ml Luria-Bertani (LB) medium plate by using an inoculating loop. Then, the bacterial suspensions in these plates were incubated to achieve a bacterial solution (~10⁹ CFU/mL) with exponential growth at 37 °C for 13 h. The obtained bacteria were diluted to 10⁸ CFU/mL, and the diluted bacterial suspension (100 µL) was evenly dispersed on LB agar plates in which CNDs with different masses (0, 0.001, 0.002, 0.004, 0.006, 0.008, and 0.010 g) had been distributed. Subsequently, these plates were irradiated by pulsed 310 nm light for five cycles (one cycle: irradiation six times within 1 min, excitation light: 310 nm, 7 mW cm⁻², each interval is 10 s). After that, they were transferred to an incubator. Similarly for Salmonella, a single colony was taken in 20 ml of LB medium at 180 rpm for 13 h to reach a bacterial concentration of 10⁸ CFU/ml. Then the same procedure as above was carried out. Finally, the colony counting method, SEM observation method, and fluorescence staining method were used to analyze the antibacterial properties of the CNDs.

Structural and optical characterization
The microstructures of all the CNDs were characterized by transmission electron microscopy (TEM, JSM-6700), and X-ray diffraction (XRD) patterns were
collected with a Bruker D8 Discover (Germany) X-ray diffractometer. For the measurement of the fluorescence and phosphorescence of the samples, the spectra were characterized using a Hitachi F-7000 PC spectrophotometer. The lifetimes were measured using an FLS-1000 spectrometer equipped with a microsecond flash lamp. The absolute PL QYs were measured using an absolute photoluminescence quantum yield FLS-1000 equipped with an integrating sphere under ambient conditions. UV–vis absorption spectra were performed using a Hitachi UH4150 UV–vis spectrophotometer. The FTIR spectra of the samples were recorded using a Bruker VERTEX-70 FTIR spectrometer. The fluorescence imaging of bacteria was performed by using a Leica TCS SP8 STED 3X laser scanning confocal microscope.

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Author contributions
K-KL conceived the idea; C-XS, LD, and K-KL supervised and coordinated all aspects of this project. S-Y.S. carried out the synthesis and characterization, and S-Y.S. and K-KL wrote the paper. S-Y.S. measured the time-resolved spectra. X-M. did the theoretical calculation. W-B.Z. and W-Y. did biological and antibacterial experiments. Q-G, Y-C-L, J-H-Z., and Q-L commented on the paper, and all of the authors discussed the results.

Conflict of interest
The authors declare no competing interests.

Supplementary information
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