



Understanding and manipulating luminescence in carbon nanodots



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ABSTRACT

Carbon nanodots (CDs), a new star in the carbon nanomaterials family, have been demonstrated to show strong luminescence, and can meet the needs of large-scale production for biological and medical applications due to their low toxicity and biocompatibility. However, their luminescence mechanisms, such as color tuning and strong excitation-dependent luminescence, are still unclear. Herein, we present a state-of-the-art understanding and manipulating luminescence in CDs by changing their environmental states and using multiple spectroscopic methods as well as the first-principles theoretical calculations. Our study reveals that the edge-carbon atoms and incorporated nitrogen atoms play critical roles in the luminescence mechanisms of CDs, and thus paves the way for manipulation of luminescence in CDs.

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1. Introduction

Luminescent carbon nanodots (CDs) are a new star member in the family of carbon nanomaterials because they possess distinct advantages, such as multicolor emissions, photo stability, low toxicity, biocompatibility and feasible large-scale production [1–6]. Recently, CDs have attracted considerable interest due to their wide application potential in bioimaging, drug delivery, sensors, and light-emitting devices [7–16]. Still, their luminescence mechanisms, especially excitation wavelength dependent photoluminescence (PL) and self-quenching of luminescence in aggregated solid state, are unclear [17–19], although a variety of models have been proposed to interpret the phenomena [20–25].

In general, quantum confinement effect, surface traps and edge states are considered as the key factors in the luminescence mechanisms of CDs [20,24]. For size-dependent PL, the quantum confinement effect obviously plays a critical role, and has been firmly demonstrated by preparing CDs with different sizes. But, PL spectra of single CDs are found to be size independent [4,26]. On

the other hand, continuous surface defect states have been suggested to be responsible for tunable emissions [19,20,27]. This widely adopted surface-defect model provides a good explanation to the excitation dependent luminescence. Very recently, distinct edge states induced by several carbon atoms on the lattice edge of CDs and so-called functional groups have been attributed to the physical origin of the common green emission of CDs and graphene quantum dots [28]. However, the edge state model seems difficult to interpret the largely tunable luminescence from blue to red commonly observed in CDs [29].

The great application potential in a wide variety of fields and the lack of a deep insight into the luminescence mechanism jointly push us to conduct this study, in which we show that the defect states and edge states induced by the doped nitrogen atoms and carbon atoms on the edge of carbon sheet, respectively, act as luminescent centers for the colorful luminescence in CDs.

2. Experimental

2.1. Synthesis

The CDs were synthesized from urea and citric acid using microwave-assisted method. Detailed description on the synthesis procedures can be referred to previous studies [13,30]. Green and

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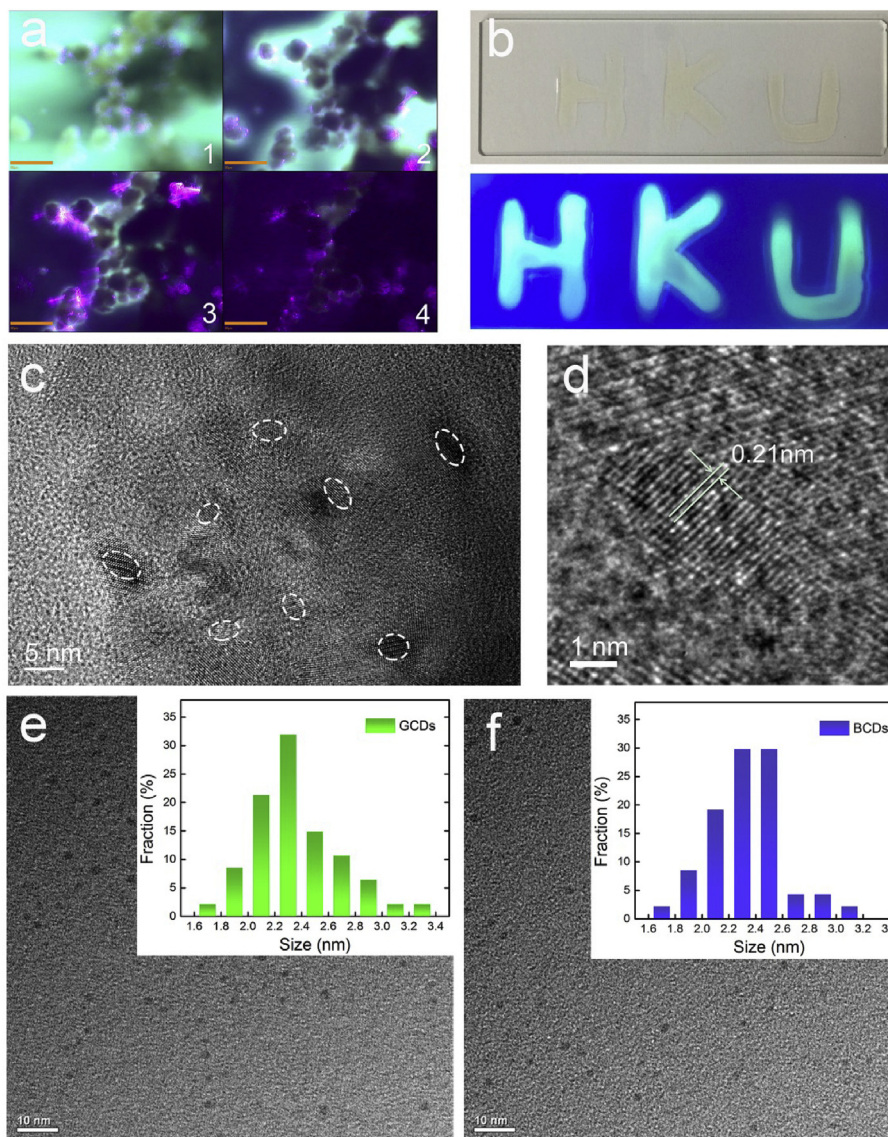


Fig. 1. (a) Optical images of green CDs dissolved in ethanol solution under the excitation of a 405 nm laser at room temperature. Green luminescence quenching can be seen as the evaporation of ethanol occurs (From 1 to 4). The scale bar is $80\mu\text{m}$ in the photographs. (b) Photographs of green CDs dispersed in PVA in solid state on a glass substrate under the illumination of the sunlight (upper) and a 405 nm laser (bottom), respectively, at room temperature. (c) and (d) HRTEM images of green CDs. The scale bars in (c) and (d) are 5 nm and 1 nm, respectively. TEM images and size distributions of (e) green CDs and (f) blue CDs. The scale bars in (e) and (f) are 10 nm. (A colour version of this figure can be viewed online).

blue CDs were prepared from urea and citric acid in different mass ratios of 2:1 and 0.2:1, respectively. Then the CDs can be added into poly (vinyl alcohol) (PVA, J&K, average M.W.: 95000) aqueous solution (0.5 g PVA in 10 mL DI water), and dried for luminescent gel. The preparation method of incorporating green CDs into NaCl crystals was described elsewhere [31].

2.2. Characterization

For the PL spectroscopic measurements, a home-assembled PL setup consisting of a Spex 750 M monochromator and a Hamamatsu R928 photomultiplier detector was used. The 325 nm laser line of a Kimmon He–Cd laser was employed as the excitation light source in the steady-state PL spectral measurements. For the variable-temperature spectroscopic measurements, the samples were mounted on a cold finger of a Janis closed cycle cryostat providing a varying temperature range from 4 K to 300 K. Room-

temperature IR spectra were registered on an IR Affinity-1 spectrophotometer. The excitation wavelength dependent spectroscopic measurements and TRPL decaying traces were obtained by using a spectrograph setup produced by Horiba PTI. The excitation light source was a 75 W Xe-lamp for the excitation wavelength dependent PL spectra. The absorption spectrum was acquired as the ratio of two synchronous scanning with and without the sample in an integrating sphere. The TRPL signal was detected by using a laser strobe method under the excitation of a pulsed nitrogen laser whose emission wavelength was 337 nm, pulse width 800 ps, and repetition frequency 10 Hz. Transmission electron microscopy (TEM) images were recorded with a FEI-TECNAI G2 F30 transmission electron microscope operating at 200 kV.

2.3. Theoretical calculations

The first-principles calculations based on the density functional

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