ABSTRACT

Correlative super-resolution microscopy reveals preferential localization of quantum defects formation on carbon nanotubes

Semiconducting single-walled carbon nanotubes (SWCNTs) exhibit excitonic fluorescence in the short-wave infrared (SWIR) region. Covalent functionalization with organic color centers (CCs) also called quantum defects or luminescent defects can modify their excitonic behavior, creating new radiative pathways that shift the emission to longer wavelengths, enhance nanotube brightness, and enable excitation through the first-order excitonic transition. This makes them unique for a wide range of applications, from deep-tissue single-particle tracking in the biological window to quantum information. However, the functionalization reaction of SWCNTs with CCs is still poorly understood, stochastic, and therefore difficult to control. To address this challenge, we employed single-molecule localization microscopy (SMLM) to reveal single CC implantation along individual SWCNTs, determining the position of each CC with nanometric precision. A custom-built multi-color super-resolution microscope was developed to simultaneously image both the SWCNT backbone (emission at 985 nm) and CCs (emission ≥ 1160 nm), ultimately enabling *in situ* monitoring of the functionalization reaction.

Examination of the spatial distribution of CCs along the nanotube axis revealed distinct preferential sites of CC formation. The positioning of the CCs on the nanotube skeleton nanotube clearly shows , a pronounced accumulation emerged near the central region. Simulations based on randomized CC distributions confirmed this non-uniformity, showing that CC formation is not random but instead biased toward preferential location. Furthermore, statistical analysis demonstrated a clear tendency for CCs to cluster spatially, suggesting that new CC preferentially form near pre-existing ones, likely driven by local strain accumulation or enhanced chemical reactivity in those regions. These findings provide direct experimental evidence that CC formation dynamics are governed by intrinsic structural and energetic heterogeneities, paving the way toward controlled engineering of quantum defects for optimized near-infrared nano emitters in biological imaging and quantum technologies.

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