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Optical studies of the growth kinetics of individual carbon nanotubes

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Introduction

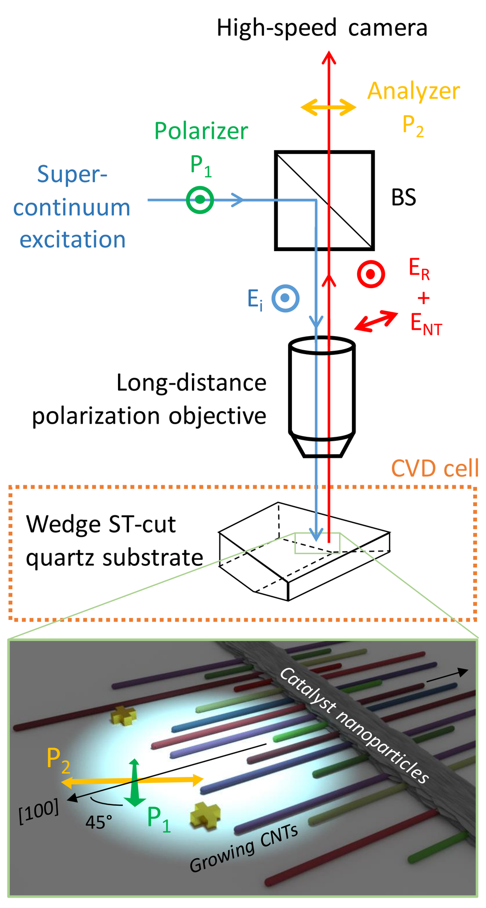
Carbon nanotubes (CNTs) are quasi one-dimensional crystals whose structure and features makes them interesting objects for optical and electronic applications. Today there are several problems on the way of its practical utilization such as necessity of narrow distribution of chiralities or high yield of given conductivity of nanotubes.

There are mainly two different methods to obtain CNTs with appropriate properties. The first type of approach is based on separation of tubes after their synthesis while the second one relies on the direct synthesis of CNTs with controlled structure.

This work addresses the second approach and aims at understanding nanotube growth mechanisms to develop selective synthesis techniques.

For this aim we use a unique homodyne polarization microscopy setup based on a principle developed at the university of Berkeley a few years ago [1] . Our setup allows us to obtain videos of individual nanotubes during CVD [2] synthesis on a substrate and at ambient pressure with a time resolution as good as 40 ms.

Homodyne polarization microscopy [3]



$$C = \frac{I - I_0}{I_0} = \frac{(1+r)^4 |A|^2}{4r^2\delta^2} + \frac{(1+r)^2 Im(A)}{r\delta}$$

Rayleigh scattering — Interference of fields

C – optical contrast

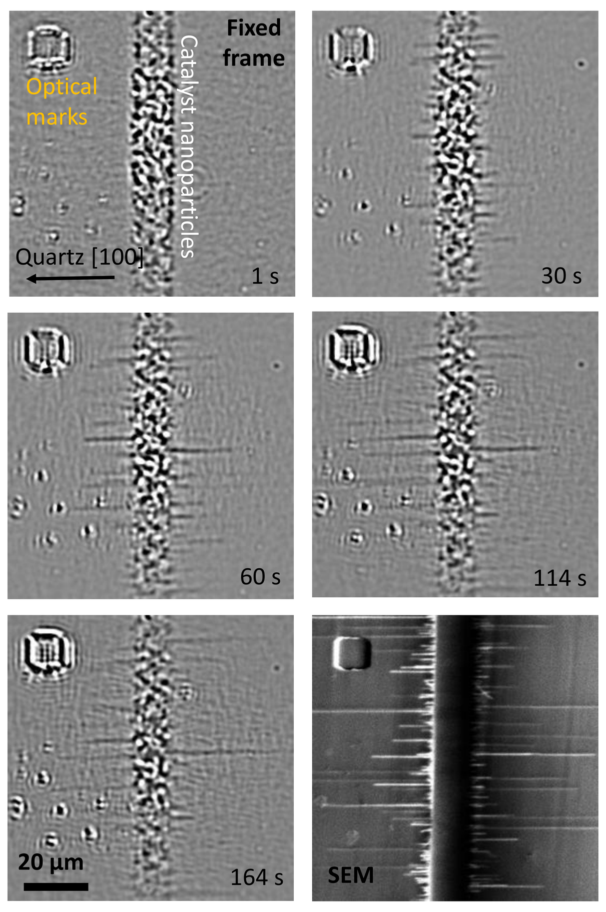
I – intensity of light from CNT

I_0 – intensity of light outside CNT

r – reflection coefficient of substrate

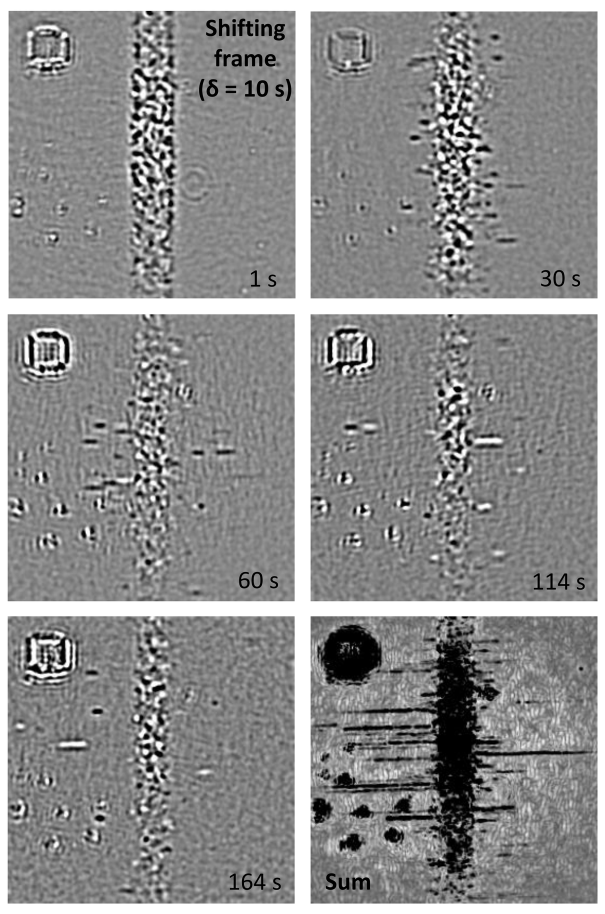
δ – deviation of angle from 90°

A – nanotube susceptibility

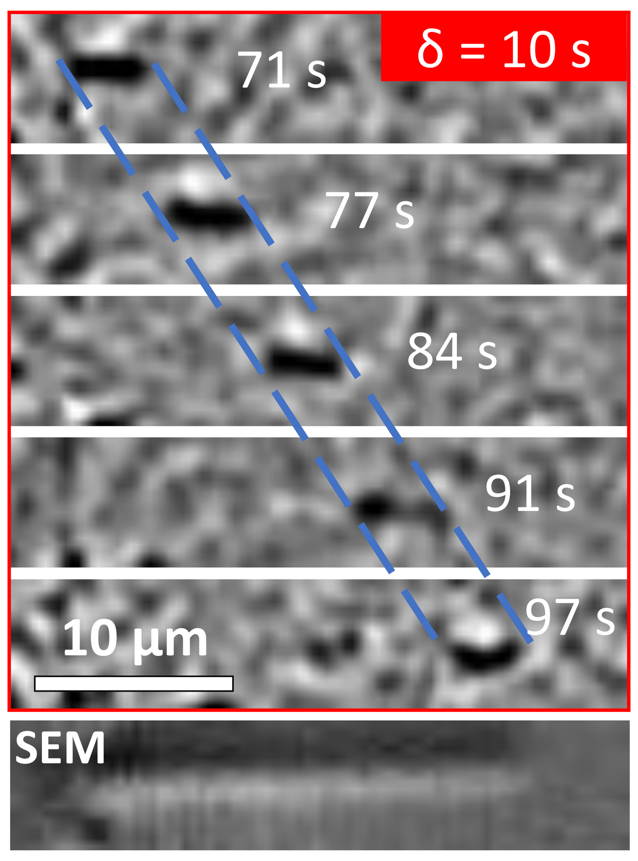


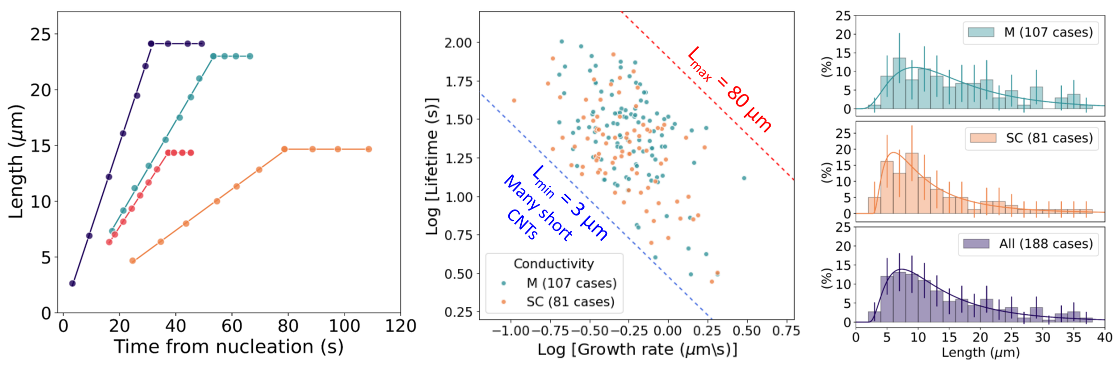
Snapshots on the left are from the videos obtained by polarization microscopy. Dark horizontal segments are nanotubes growing from that thick vertical catalyst line that was patterned on the substrate by UV lithography. From such videos we can get the information about time of growth beginning and end, calculate lifetime and extract data about tube's growth rate.

Snapshots on the right are from video that was processed with developed by our team shade correction technique that allow us to got the date appeared on video during fixed time period. Such treatment allow to resolve tubes in time, increase contrast and define tubes that are grown and pass through structural changes.



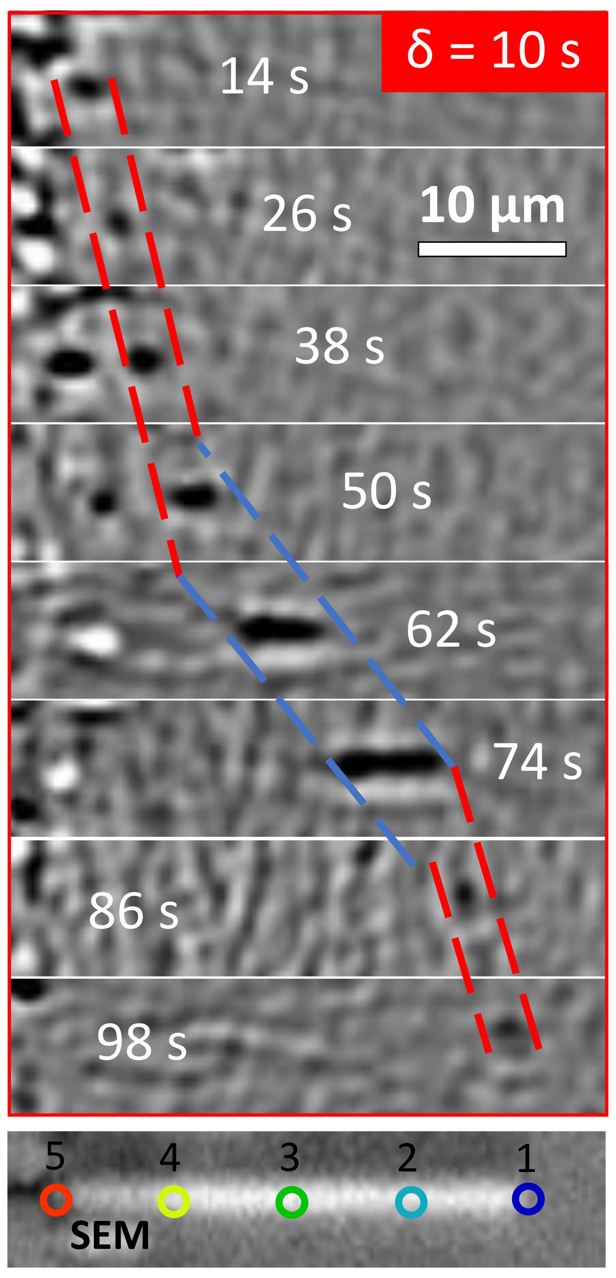
Linear Growth

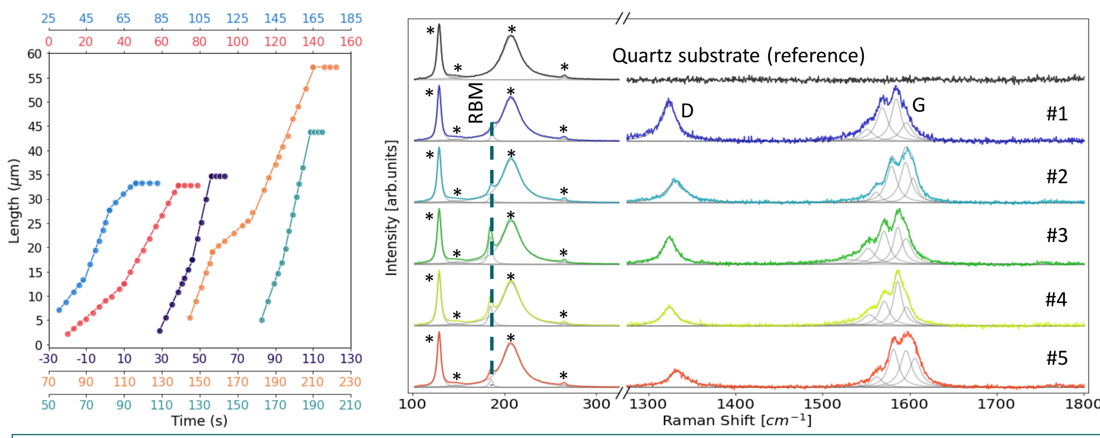




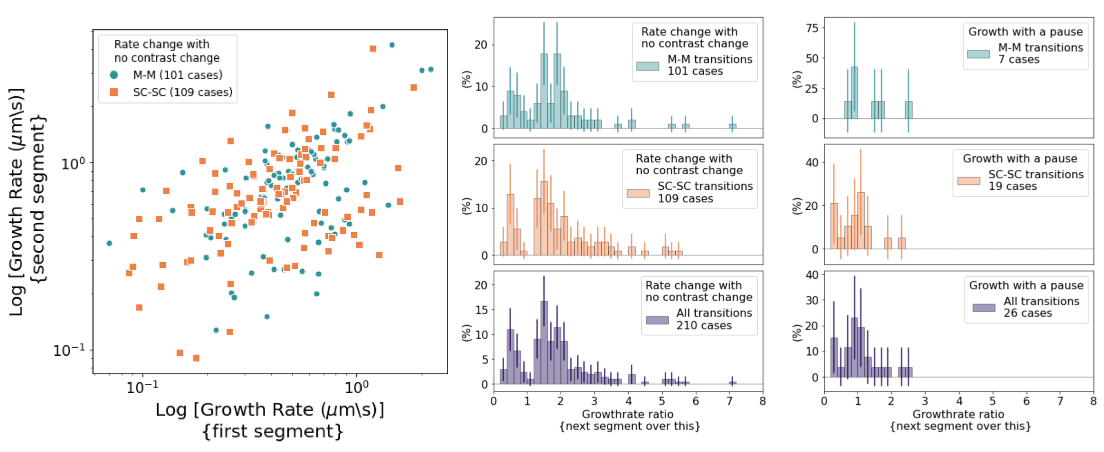
We evidenced tubes growing with constant speed from beginning to the end. During these linear growths, the OI contrast, the SEM contrast and the Raman signatures remain constant all along the tube. Note that, contrary to past reports, we never observed an exponentially decreasing growth rate at the level of an individual tube: an exponential law emerges only when summing up a large number of individual events, exactly like radioactive decay (see the top left figure).

Change of growth rate without change of structure

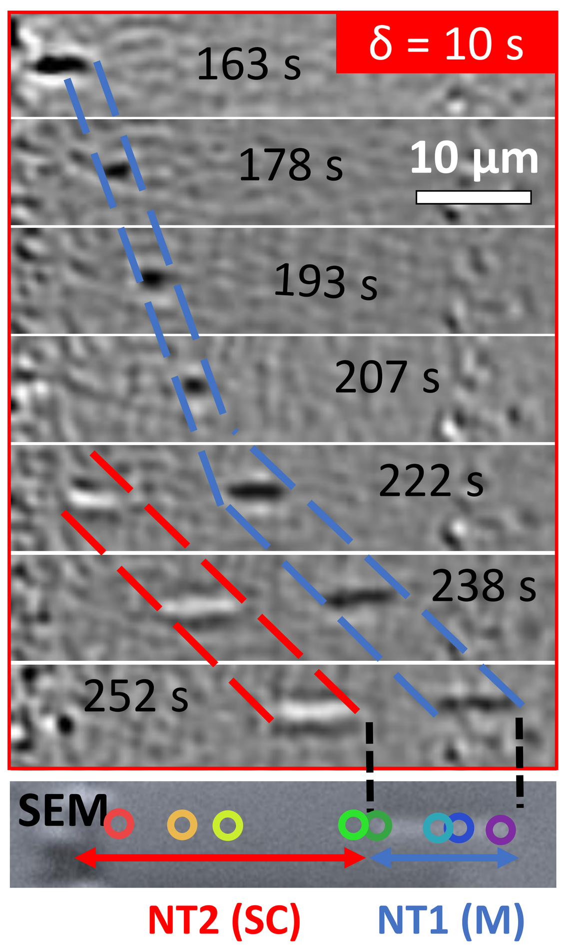


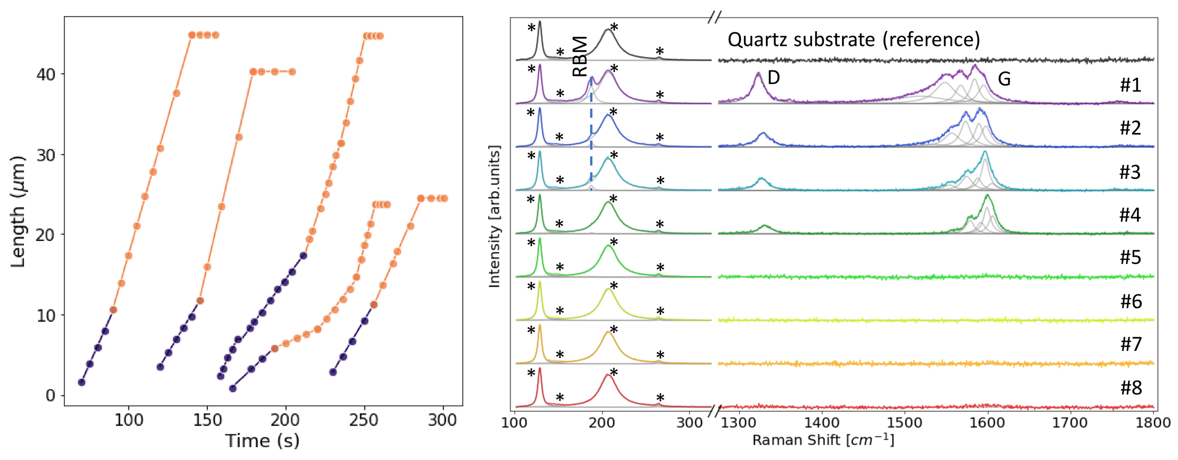


Another type of kinetics is a change of growth rate with no apparent change of structure, neither in the OI or SEM contrast, nor in the Raman signature. On differential videos, such an event manifests only by a change of segment length during the growth. These features support that the change of rate is not related to the nanotube structure but to a discontinuous phase or structure transition of the catalyst nanoparticle.



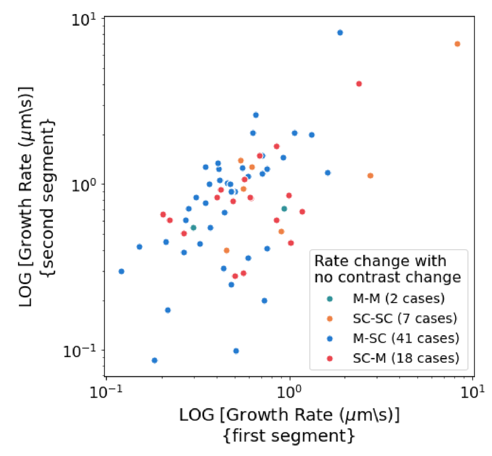
Change of growth rate with change of structure





In some cases, the change of rate is correlated with a change of nanotube structure as evidenced by either a change of OI and SEM contrasts, or by a change of Raman features along the tube.

Such event demonstrates a base-growth mechanism in agreement with other groups.



Conclusion

First evidence of complex kinetics of individual SWCNTs in real conditions

Kinetics controlled influenced significantly by stochastic changes catalyst phase

Growth rate does not depends on nanotube's metallicity type

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[1] Kaihui Liu, et al. Nature Nanotech volume 8, pages917–922(2013)
[2] Jourdain, V. & Bichara, C. / Carbon N. Y. 58, 2–39 (2013)
[3] Monniello, L. et al. Phys. Rev. B 99, 1–10 (2019)

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