

Spontaneous twisting of a collapsed carbon nanotube

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ABSTRACT

We study the collapsing and subsequent spontaneous twisting of a carbon nanotube by *in situ* transmission electron microscopy (TEM). A custom-sized nanotube is first created in the microscope by selectively extracting shells from a parent multi-walled tube. The few-walled, large-diameter daughter nanotube is driven to collapse via mechanical stimulation, after which the ribbon-like collapsed tube spontaneously twists along its long axis. *In situ* diffraction experiments fully characterize the uncollapsed and collapsed tubes. The experimental observations and associated theoretical analysis indicate that the origin of the twisting is compressive strain.

1 Introduction

Collapsed carbon nanotubes (CCNTs) [1] share the attributes of conventional (i.e., inflated or cylindrical) nanotubes from which they are typically derived as well as attributes of graphene nanoribbons, which they

closely resemble geometrically. Indeed, a collapsed nanotube may be viewed as the realization of an atomically perfect multi-layer graphene nanoribbon, in that all edge atoms are fully bonded without hydrogenation or other functionalization. CCNTs have been studied extensively, both experimentally and

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theoretically [2–11]. The physical origin of CCNTs is straightforward [1]: A conventional carbon nanotube minimizes its elastic curvature energy by assuming a cylindrical shape, but if the nanotube has a large enough diameter and few enough shells (i.e., walls), the van der Waals energy tapped by allowing the opposing “faces” of the inner walls to come into close proximity and stick together can overwhelm the increase in the strain energy at the edges, leading to a metastable or even globally stable collapsed state. A collapsed single-walled carbon nanotube resembles a bilayer graphene nanoribbon, while a collapsed double-walled carbon nanotube resembles a four-layer graphene nanoribbon, etc. Importantly, the chirality of the innermost shell of the uncollapsed nanotube severely constrains the allowed layer-to-layer stacking of adjacent inner layers in the collapsed tube.

Similar to the case of graphene nanoribbons, CCNTs can undergo additional structural changes such as curling, folding, or, rather strikingly, twisting along their long axis with a well-defined periodicity [12, 13]. Although several theories have been advanced to account for twisting in CCNTs [12, 14–16] as well as in graphene nanoribbons [17–20], there is no consensus on the dominant driving mechanism. The experimental situation is even less clear; CCNTs are often observed accidentally [12, 14] or synthesized in the solution phase (wherein the inner core is extracted by ultrasonication) [21], and there is little understanding of the dynamics of the formation/twisting process or the sensitivity of those processes to nanotube geometry. No real-time transmission electron microscopy (TEM) tracking of such processes has been reported. Deeper insight into such processes would assist in the practical application of CCNTs in nanomechanical systems [22, 23].

Here, we employ *in situ* TEM to track the collapse and subsequent spontaneous twisting of a carbon nanotube in vacuum. A nanomanipulator (Nanofactory Instrument AB) is first used to create a custom-sized nanotube in the microscope (JEOL 2010 TEM operated at 80 keV) by telescopically extracting shells from a multi-walled parent nanotube. The daughter nanotube has a diameter and wall number that make the nanotube prone to collapse, and it is induced to do so via mechanical perturbation by the nanomanipulator.

The CCNT is then singly clamped at its end, and while suspended in vacuum, it is observed to spontaneously twist along its long axis. By means of TEM imaging and electron diffraction, we track the structural changes during collapsing and twisting. We use the experimental observations to test various theoretical models for twisting and show that the main driving force for twisting of a CCNT is compressive strain.

2 Experimental

The left panel drawings in Fig. 1 schematically show our experimental process to create a CCNT *in situ* by nanomanipulation. Initially, one end of the inflated conventional parent multi-walled carbon nanotube (right side in Fig. 1(a)) is fixed via silver paint to a copper mesh TEM grid, which is then attached to a stationary sample holder in the TEM. Next, the opposite (left) end of the parent nanotube is spot-welded *in situ* via a bias voltage (typically between 1 and 4 V) applied to the tungsten tip of the piezo-controlled nanomanipulator, which serves as both a mechanical manipulator and mobile electrode [24]. As the tungsten tip moves to the left (Fig. 1(b)), it extracts one or more walls of the parent tube, yielding the daughter tube, which telescopes or slides off of the fixed core. The goal is to produce a daughter tube that is susceptible to collapse, i.e., one that has few walls and a relatively large outer diameter (limited, of course, by the outer diameter of the parent tube). When a suitably long daughter tube has been extracted (Fig. 1(c)), the collapse of the daughter tube is mechanically initiated by a small displacement of the tungsten tip transverse to the tube axis (vertical arrow in Fig. 1(c)). This typically leads to a runaway zipper-like collapsing process, wherein the daughter tube first collapses locally at the perturbation site, and the collapsed region then quickly propagates along the length of the daughter tube, until the collapse is halted by the core plug. The overall length of the collapsed daughter tube can be further and controllably extended by additional leftward movements of the tungsten tip (as the daughter tube telescopes from the stationary core, the newly exposed “hollow” region immediately collapses via the zipper effect, still driven by van der Waals forces). The maximum

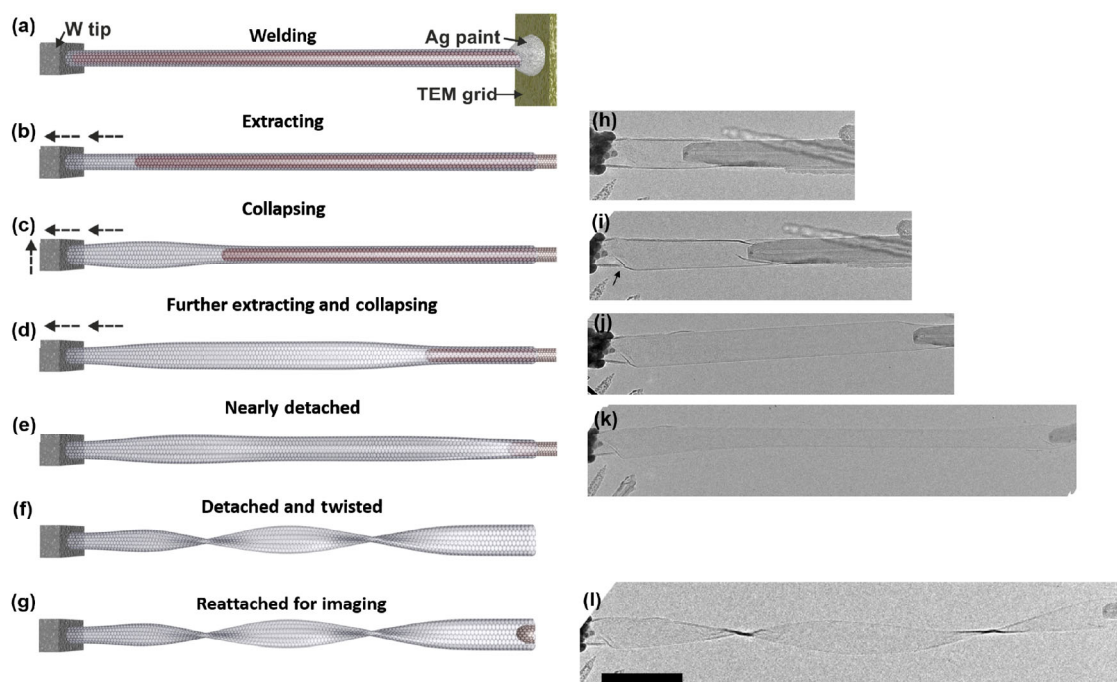


Figure 1 Schematic of our experimental setup ((a)–(g)) along with corresponding TEM ((h)–(l)) images on the right. As the W tip is moved to the left, it strips off a large diameter (26 nm) three-walled carbon nanotube that first collapses and then twists. For graphical convenience, images have been displaced laterally so that the (mobile) W tip always remains at the left edge of the image. The dashed arrows that point to the left in (b), (c), and (d) indicate the direction in which the W tip actually moves. The other dashed arrow that points vertically in (c) indicates the direction of the mechanical perturbation from the W tip. The solid arrow in (i) points to the kink formed by the mechanical deformation from the W tip. The black scale bar in (l) is 100 nm; (h)–(l) have common scale.

length of the doubly clamped collapsed daughter tube is equal to the total length of the multi-walled parent tube (Fig. 1(e)). Further movement of the tungsten tip to the left yields full detachment on the right end of the daughter tube from the core, resulting in a CCNT singly clamped at one end and suspended in vacuum at the other end. Spontaneous twisting can then occur unimpededly (Fig. 1(f)). Note that *in situ* spot welding enables reattaching the right end of the daughter CCNT to the remaining core tube or the copper mesh TEM grid itself, resulting in a doubly clamped twisted CCNT (Fig. 1(g)). Such reattachment of the free end facilitates the TEM imaging of the twisted CCNT mainly by quenching vibrations.

3 Results and discussion

The right panel of the TEM images in Fig. 1 illustrates an actual experiment performed as outlined above, with direct correspondences between the TEM images and the adjacent schematic drawings. The inflated

parent tube (visible on the right side of Fig. 1(h)) has about 31 walls, and its outer diameter is 26 nm. The tailored daughter tube has three walls (see Fig. S1 in the Electronic Supplementary Material (ESM)) and an outer diameter of 26 nm; such a daughter tube is very susceptible to collapse as it is more than three times wider than the critical radius of a three-walled tube (7.6 nm) [6]. The arrow in Fig. 1(i) indicates where the collapse of this tube was initiated via mechanical perturbation. (Note that the spot weld to the tungsten tip prevents the extreme left end of the daughter tube from completely collapsing by holding the tube open.)

The width of the daughter nanotube after collapse is 39 nm, as determined from the TEM image in Fig. 1(j), which is consistent with what is expected for the collapsed state [21]. When the CCNT is nearly completely detached from the inner core, we observe narrowing at the center (Fig. 1(k)), where the edges fold towards the tube axis. When the tube is fully detached from the inner core and is in a singly clamped condition, it twists along its axis with two nodes.

Figure 1(l) clearly shows the twisted condition (note that for Fig. 1(l), the right end of the nanotube has been reattached to the stationary core on the right to minimize the vibration for enhanced TEM characterization). We repeated the above experiments with other daughter nanotubes derived from independent parent tubes and obtained similar results, as outlined in Table 1. Corresponding TEM images are provided in Fig. S2 in the ESM.

The ability to structurally characterize a given nanotube before and after collapse is essential for elucidating why CCNTs twist. We analyze the lattice registry of the tailored CCNT in Fig. 1 before and after twisting by selected area electron diffraction (SAED). The SAED pattern in Fig. 2(b) was taken at the right end of the CCNT, where it is partially collapsed and partially tubular (outlined by a blue circle in Fig. 2(a)). The pattern shows only two sets of six-fold graphitic diffraction patterns corresponding to the two opposing graphitic walls of the nanotube.

This indicates that all three walls of the daughter nanotube have the same chirality (see Fig. S3 in the ESM). The streaked features in the diffraction pattern are due to the curvature of the inflated part of the tube, while the more defined round diffraction spots (near the streaks) come from the flat region of the collapsed part. Because the graphitic diffraction spots for the inflated and collapsed parts are nearly identical, we conclude that the opposing faces of the inner walls maintain their orientation during collapse. The numerical value of the chiral angle for the inflated daughter tube is $\theta = 27.1^\circ \pm 0.5^\circ$ [25], which is consistent with the relative rotation angle between the graphitic layers of the collapsed part ($\beta = 7^\circ \pm 0.5^\circ$). Note that the diffraction pattern taken at the center of the collapsed part (before twisting, Fig. 1(j)) indicates

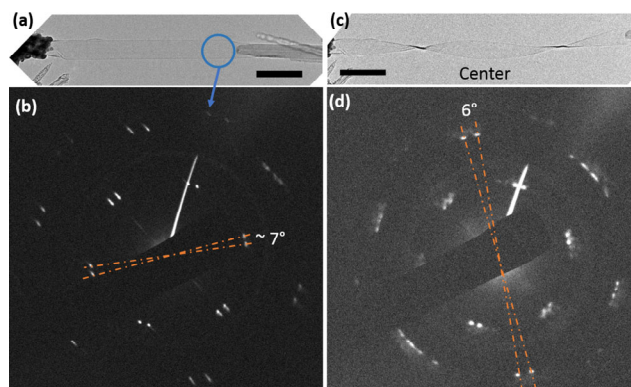


Figure 2 TEM image of the CCNT before twisting (a) and SAED pattern (b) of a partially collapsed region outlined by a blue circle (i.e., the left side of the circle is collapsed, and the right side is not). TEM image after twisting (c) and SAED pattern (d) taken from the central part of the tube. The scale bars in (a) and (c) are 100 nm.

the same relative rotation angle between graphitic layers (see Fig. S4 in the ESM). More importantly, we find that the orientation is nearly unchanged even after twisting, as demonstrated by the SAED pattern (Fig. 2(d)) obtained from the center of the twisted CCNT (Fig. 2(c)), which gives nearly the same orientation ($\beta \sim 6^\circ$) as that before twisting ($\beta \sim 7^\circ$).

We now examine the registry [12, 14, 15] and edge [17] mechanisms for the twisting of a CCNT. In the registry mechanism, twisting in a CCNT is driven by a lattice registry effect between opposing walls. Attaining the most stable energy configuration drives the local lateral shifting of the opposing walls of the CCNT, which then manifests itself as twisting along the axis of the CCNT. The edge mechanism for twisting is adapted from mechanical stability studies of a freestanding single-layer graphene nanoribbon [17], or analogously, from the well-known instabilities of macroscopic sheet metal strips processed via rollers [26, 27]. Here, twisting is attributed to non-uniform stress in the ribbon resulting from the elongation of the ribbon edge material relative to the center material. In graphene nanoribbons, the length differential originates from the lattice reconstruction of the edge atoms [17, 18], while for sheet metal, it results from non-planar rollers that non-uniformly elongate the material [26]. As we demonstrate below, neither the registry nor the edge mechanism accounts for our observations, and we present an alternate model.

Table 1 Experimental data (number of walls “ n ”, tube width “ W ”, and twist period “ T ”) and calculated strains “ ε ” for the examined CCNTs in Fig. 1 and Fig. S2 in the ESM

Tube	n	W (nm)	T (nm)	ε (%)
i (Fig. 1)	3	39.0	600	1.30
ii (Figs. S2(a) and S2(b))	3	7.0	400	0.09
iii (Figs. S2(c) and S2(d))	5	18.6	536	0.30
iv (Figs. S2(e) and S2(f))	1	31.5	1,592	0.11



Our theoretical examination of competing mechanisms is derived from a continuum elasticity model, which we justify using first-principles density functional theory (DFT) calculations using the Quantum ESPRESSO package [28]. First, we simplify our calculation by showing that the bulging edges of a typical CCNT have very little effect on its overall elastic behavior. We justify this simplification by two DFT calculations of the elastic properties relevant for twisting: First, we calculate the energy cost per atom of uniaxial compression for pristine graphene and for an armchair (40, 40) CCNT, and we find these two energy costs to agree within 1% in a range of strain from -1.5% to $+1.5\%$. After the full structural relaxation of a CCNT, we find that the lattice constant along the nanotube axis is reduced by only 10^{-4} relative to the pristine graphene sheet (namely, $2.4312 \text{ \AA}$ in the pristine graphene sheet and $2.4309 \text{ \AA}$ in the CCNT).

A second quantity we examine is the softening of the out-of-plane (flexural) phonon mode by uniaxial compression. As found in Ref. [29], this phonon softening mechanism causes pristine graphene sheets to buckle. We find that flexural phonon softening is only slightly different (by less than 20%) in graphene than in a CCNT. The discrepancy here is larger than in the first case likely because we approximate the flexural phonon eigenvector in a CCNT in order to render the calculation manageable¹. Therefore, in the following analysis of the CCNT twisting, we will temporarily ignore the effects of the bulges and treat the CCNTs as a stack of flat, pristine graphene sheets. Later, we will estimate the error caused by this modification.

The elastic properties of pristine graphene sheets (and therefore of CCNTs) are well described by a continuum model [30]. Using the lowest order of the elastic continuum model, we obtain (see the ESM) the following areal energy density (E) of a twisted CCNT with width W and twist period T

$$E = \frac{\pi^2}{6} \varepsilon C \left(\frac{W}{T} \right)^2 + \frac{\pi^4}{40} C \left(\frac{W}{T} \right)^4 \quad (1)$$

¹ Instead of computing the entire dynamical matrix of a CCNT, we computed a single matrix element assuming a simple form of a flexural phonon, where all atoms in the CCNT move perpendicularly to the collapsed plane of the CCNT.

where the only material-dependent quantity is the elastic modulus C , which we find to be equal to $23 \text{ eV/\AA}^2$ in both graphene and CCNTs (within 0.2%). Our calculated value of C is in good agreement with previous calculations by Yakobson et al. [31]. The parameter ε in Eq. (1) represents a strain along the long nanotube axis, either due to stress or charge doping. (The effects of stress and doping are indistinguishable in the continuum model.)

We use Eq. (1) to calculate the elastic energy cost of twisting. Inserting the parameters for the tailored daughter nanotube in Fig. 1, i.e., $W = 39 \text{ nm}$ and $T = 600 \text{ nm}$, into the second term (and setting ε equal to zero) shows that twisting of the CCNT costs only 2.8 meV per carbon atom. In the registry mechanism proposed in Refs. [14, 15], this increase in the elastic energy is compensated by a change in the lattice registry between the opposing graphitic walls of the CCNT. However, as discussed earlier, our *in situ* TEM study shows nearly no change in the relative rotation angle between the opposing walls upon twisting (namely, it is 7° before and 6° after twisting). There is no substantial energy difference between these two lattice mismatch angles because in both cases, the lattice registry is an equal admixture of AA and AB stacking. Even if we consider an extreme case where the interlayer interaction changes from $\beta = 7^\circ$ all the way to the AB stacking (i.e., $\beta = 0^\circ$) we estimate that the energy reduction is at most only 0.8 meV per atom, more than three times smaller than the change in the twisting elastic energy. Therefore, we conclude that lattice registry is not responsible for twisting the CCNT. (This estimate takes into account the fact that out of the three carbon nanotube walls, only the innermost interface exhibits a change in the lattice registry. It also accounts for the energy difference between AA- and AB-stacked graphite, which is 10 meV per carbon atom.)

We now turn to the edge mechanism, i.e., the possibility that the edges of the CCNT behave differently than the bulk, resulting in, for example, a differential elongation and consequent rippling and/or twisting. First of all, the CCNT in Fig. 1 is quite large (39 nm wide), and only about 5% of its carbon atoms are at the edge of the tube; hence, the energy reduction at the edge would have to be quite large (56 meV per

atom) to compensate for the energy increase due to the twist. Therefore, although this mechanism may be appropriate for describing rippling/twisting in narrow graphene nanoribbons, it is ruled out for twisting in our typical CCNTs. Second, because the structural relaxation of CCNT gives a slightly compressed lattice constant (by 10^{-4}) relative to the pristine graphene sheet, we expect the edges of the CCNT to oppose twisting deformation. This is in contrast to the case of the bare-terminated edges of a graphene nanoribbon, which expand and can thus induce twisting [32].

We now specifically discuss our proposed mechanism for CCNT twisting. An important feature of Eq. (1) is that the restoring energy cost of the twisting is proportional to the fourth power of W/T , while the effect of the strain ε due to external loading is proportional to the second power of W/T . Therefore, within our model, even a slightest negative ε value will twist a CCNT. In other words, the total energy E is minimized for a finite twist period T satisfying the following relation (see the ESM).

$$\frac{W}{T} = \frac{\sqrt{10}}{\pi\sqrt{3}} \sqrt{-\varepsilon} \quad (2)$$

This relation is graphically represented by a heavy purple line in Fig. 3. The insets in Fig. 3 show the

energy density (E) of a CCNT vs. the W/T ratio under both compressive (left graph) and tensile (right graph) strain.

We now include the correction to Eq. (2) due to the bulged edges of a CCNT. While the energy penalty for twisting a graphene sheet is proportional to the fourth power of W/T , the penalty originating from a non-perfect edge termination gives an energy cost that is only proportional to the second power of W/T . Therefore, non-perfect edge-termination would result in a non-zero critical strain, below which the CCNT twists. While this effect is important in a bare-edged narrow graphene nanoribbon [32], it is small in the case of a wide CCNT. From our structural relaxation of a CCNT and following Ref. [31], we estimate that the critical strain for a 39 nm wide CCNT is only 6.0×10^{-5} .

Inserting W (39 nm) and T (600 nm) into Eq. (2), we find that a compressive strain of 1.3% produces the observed twisting of the CCNT in Fig. 1 (Tube *i* in Table 1). The complementary compressive strains for the other experimentally examined twisted CCNTs are calculated in the same manner and are also presented in Table 1. The strains range from 0.09% to 1.3%.

In a typical collapsing/twisting scenario occurring in the nanotube synthesis chamber, non-local stresses induced by extreme temperature gradients, collisions with high-energy ions, gas flow, etc., are likely a

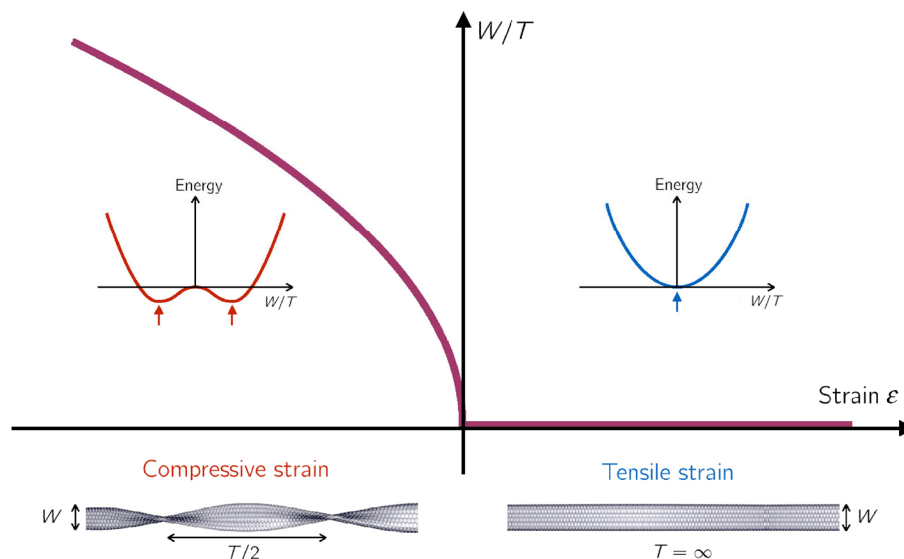


Figure 3 Our continuum model predicts that the CCNT will twist under an infinitesimal compressive strain $\varepsilon < 0$. The two small inset graphs show the dependence of the energy (E) on the ratio of the CCNT width (W) and the twist period (T) both under compressive (red graph) and tensile strain (blue graph). The purple line shows the ratio W/T from Eq. (2), which minimizes the energy of the CCNT.

common occurrence, which accounts for “naturally occurring” twisted CCNTs [1]. In our more controlled, clamped-free CCNT configuration in Fig. 1, a strain ε can result from both internal and external forces, including compression caused by a charge imbalance [33]. Charge imbalance can result from intrinsic doping (common to multi-walled carbon nanotubes due to the presence of defects or guest atoms from impurities in the growth chamber) [34, 35] and possibly to some degree from the TEM imaging beam itself.

Equation (2) reveals that the twist period T of a CCNT is proportional to W and inversely proportional to $\sqrt{\varepsilon}$. The available range of T is thus from a maximum of ∞ (for zero strain) to a minimum dictated by the width W of the CCNT and the maximum strain $\varepsilon_{\max}$ sustainable by the CCNT without catastrophic wrinkling. Taking $W_{\min} = 2$ nm (the smallest possible CCNT), and as a rough estimate, $\varepsilon_{\max} \sim 2\%$, we obtain $T_{\min} = 24$ nm [6].

Interestingly, while the continuum model predicts that a CCNT will twist even under an infinitesimal external load (see Fig. 3), previous work in Ref. [29] predicts that a related distortion, i.e., the rippling of a graphene sheet, occurs only when the load is above some critical value.

4 Conclusion

In conclusion, we find that compressive strain, regardless of how small, causes a CCNT to twist along its long axis. The results of *in situ* TEM experiments revealed that the interaction of the graphitic walls (i.e., the lattice registry effect) and in general, the chirality of the carbon nanotube have negligible effects on CCNT twisting. Our results provide insights into the dynamics of carbon nanotubes during the collapsing/twisting process, and as such, they are valuable for designing nanoscale actuators [22, 23], springs, inductors, etc., for nanoelectromechanical systems.

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