

Supplementary Information

Confined ionic order in atomic nanowires of rare-earth chlorides unveiled via symmetry-guided structural screening

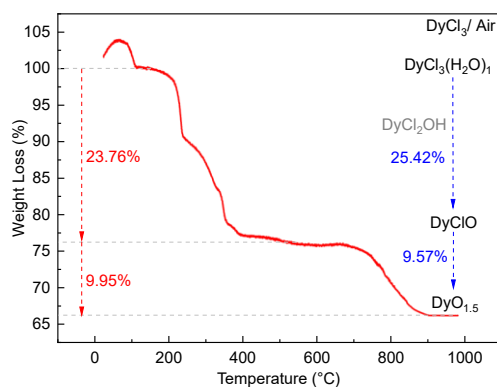
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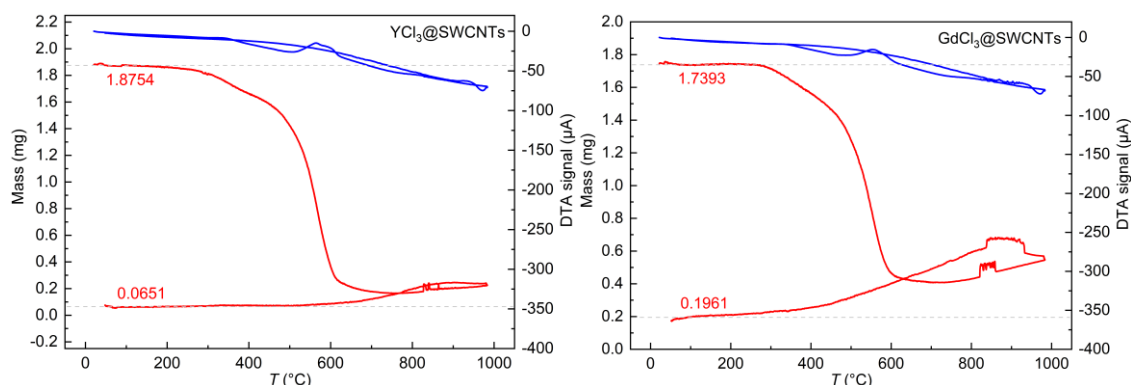
0 Table of contents

0	Table of contents	2
1	Thermogravimetric analysis (TGA)	3
2	STEM & TEM	4
3	Guessing metal atom positions from the STEM-HAADF images	10
3.1	Type "X" (TX)	10
3.2	Type "Zigzag" (TZ)	10
3.3	Type "Staggered" (TS)	12
3.4	Type "Bamboo" (TB)	13
3.5	Type "helix" (TH)	14
3.6	Type "U" (TU)	16
3.7	Type "three lines" (T3)	17
3.8	A short Summation of guessing configurations	18
4	Exploring the structure	19
5	XAFS	25
5.1	DyCl ₃	25
5.2	Quenched DyCl ₃	25
5.3	DyCl ₃ @SWCNTs	26
5.4	ErCl ₃ @SWCNTs	26
5.5	YCl ₃ @SWCNTs	27
6	X-ray photoelectron spectroscopy (XPS)	28
7	Raman spectroscopy	31
8	Magnetic properties	32
8.1	Crystalline DyCl ₃ & Quenched DyCl ₃	32
8.2	DyCl ₃ @SWCNTs	35
8.3	ErCl ₃ @SWCNTs & GdCl ₃ @SWCNTs	38
8.4	CASSCF calculation	41
9	Conductivity	44
10	Electronic band structure and charge transport properties	46
10.1	The Y ₆ Cl ₁₈ @SWCNT(14,7) model	46
10.2	The deformed SWCNT(14,7) model	47
10.3	The pristine SWCNT(14,7) model	47

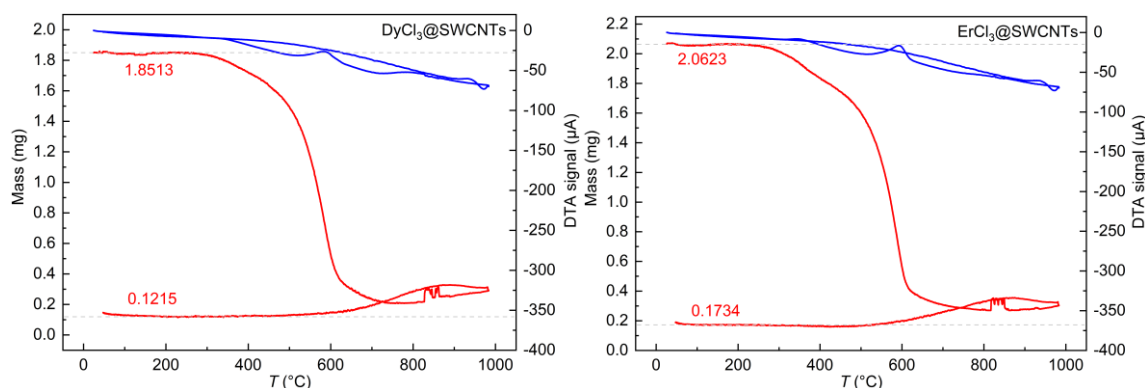
1 Thermogravimetric analysis (TGA)



Supplementary Figure 1. The TGA curve of DyCl_3 under air atmosphere. DyCl_3 is oxidized to Dy_2O_3 under an air atmosphere; similar behavior is expected for other LnCl_3 species.



Supplementary Figure 2. The TGA curve (red) and the differential thermal analysis (DTA) curve (blue) of $\text{YCl}_3\text{@SWCNTs}$ (left) and $\text{GdCl}_3\text{@SWCNTs}$ (right) under air atmosphere.



Supplementary Figure 3. The TGA curve (red) and the differential thermal analysis (DTA) curve (blue) of $\text{DyCl}_3\text{@SWCNTs}$ (left) and $\text{ErCl}_3\text{@SWCNTs}$ (right) under air atmosphere.

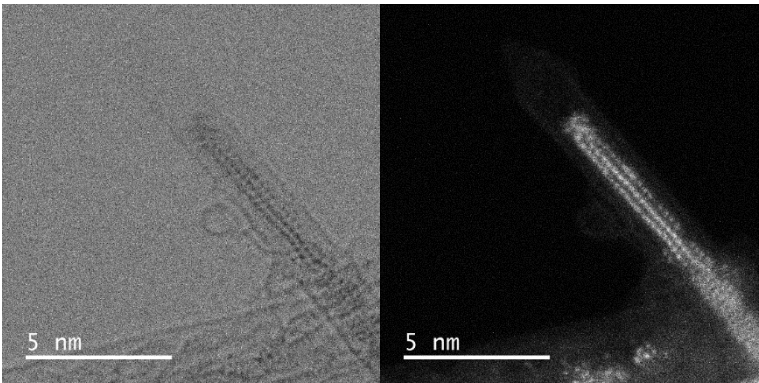
Supplementary Table 1. Mass fraction of encapsulated LnCl_3 determined from thermogravimetric analysis.

	Sample Weight (mg)		n_{Ln} (μmol)	m_{LnCl_3} (mg)	m_{SWCNTs} (mg)	$m_{\text{LnCl}_3}/m_{\text{total}}$
	Initial	End				
YCl_3	1.8754	0.0651	0.5766	0.1126	1.7628	6.00%
GdCl_3	1.7393	0.1961	1.0819	0.2852	1.4541	16.40%
DyCl_3	1.8513	0.1215	0.6515	0.1752	1.6761	9.46%
ErCl_3	2.0623	0.1734	0.9066	0.2481	1.8142	12.03%

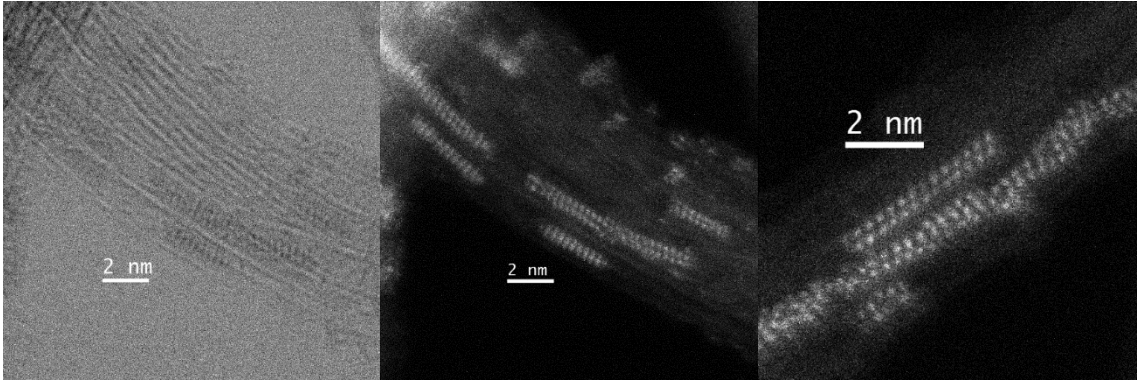
2 STEM & TEM

Supplementary Table 2. Nanowire structures in $\text{LnCl}_3\text{@SWCNTs}$ ($\text{Ln} = \text{Y}, \text{Gd}, \text{Dy}, \text{Er}$) observed by STEM.

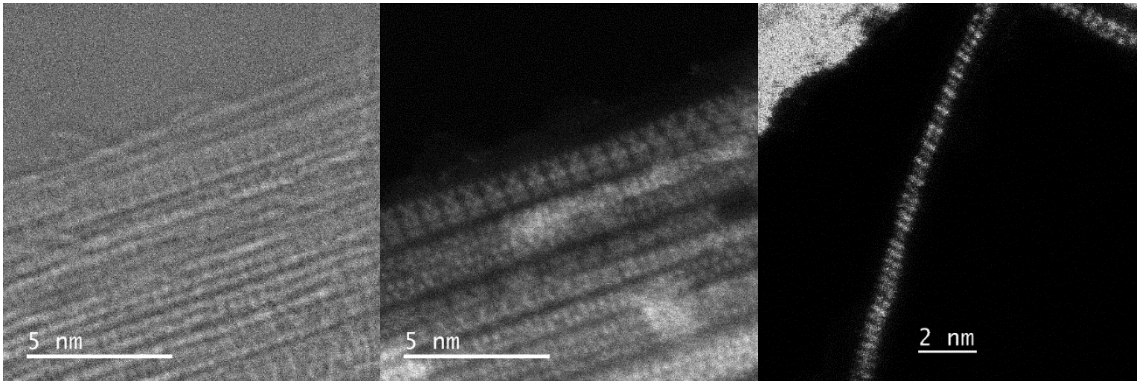
Systems	Types of nanowires
$\text{YCl}_3\text{@SWCNTs}$	N/A, STEM measurements were not performed.
$\text{GdCl}_3\text{@SWCNTs}$	TZ; poorly ordered
$\text{DyCl}_3\text{@SWCNTs}$	TB; TS; TU; TX; poorly ordered
$\text{ErCl}_3\text{@SWCNTs}$	TX; TH; poorly ordered



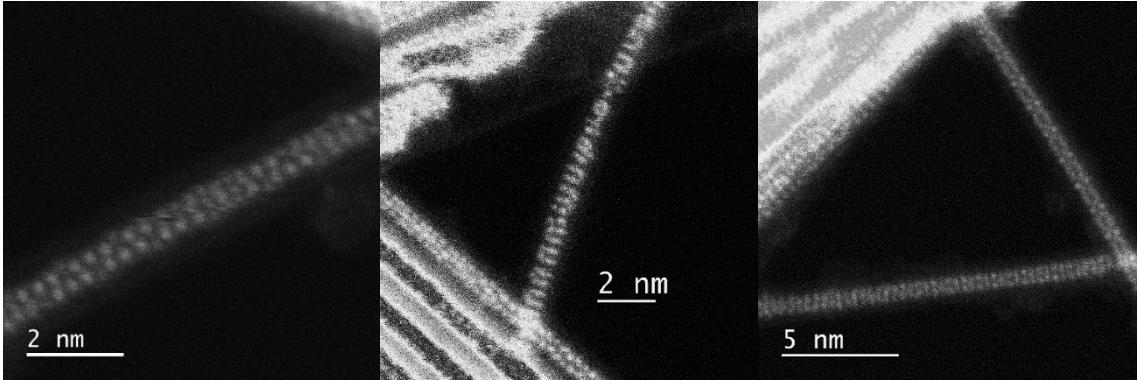
Supplementary Figure 4. ABF image (left) and HAADF image (right) of $\text{GdCl}_3\text{@SWCNTs}$.



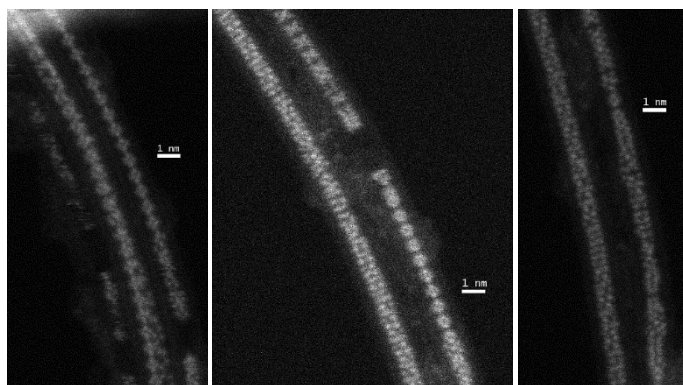
Supplementary Figure 5. ABF image (left) and HAADF images (middle, right) of $\text{GdCl}_3\text{@SWCNTs}$.



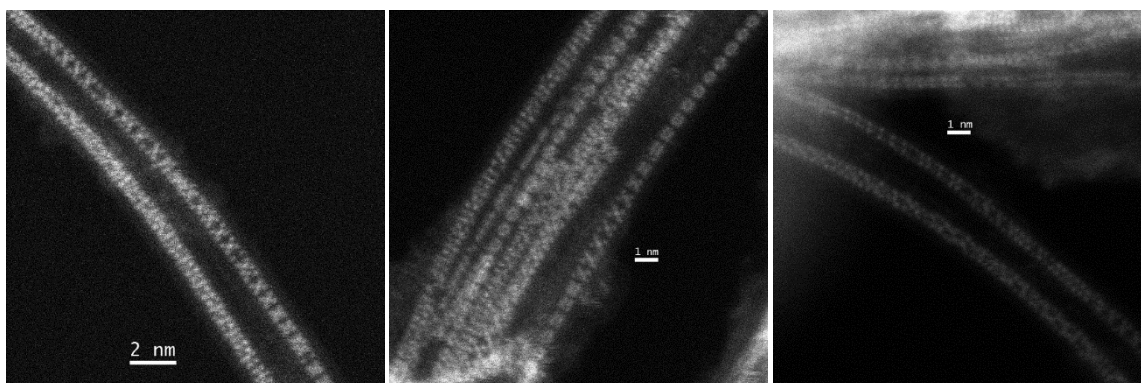
Supplementary Figure 6. ABF image (left) and HAADF images (middle, right) of $\text{DyCl}_3\text{@SWCNTs}$.



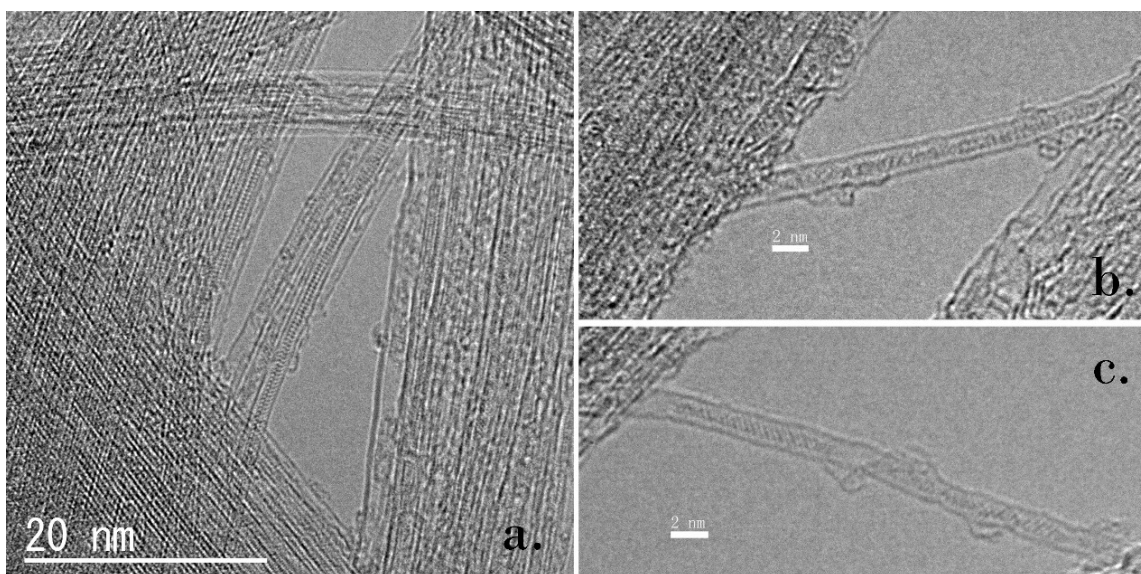
Supplementary Figure 7. HAADF images of $\text{DyCl}_3\text{@SWCNTs}$.



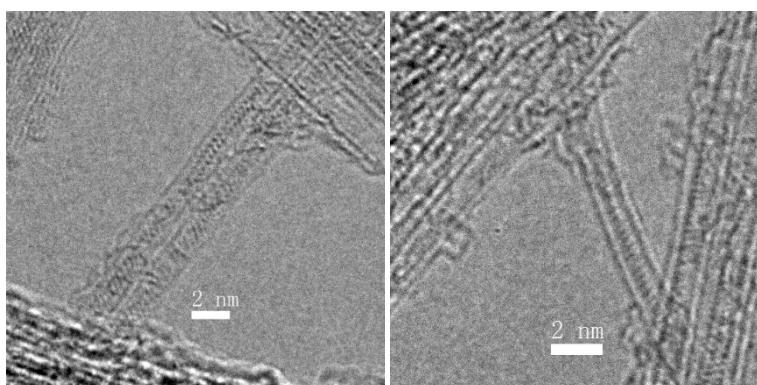
Supplementary Figure 8. HAADF images of ErCl₃@SWCNTs.



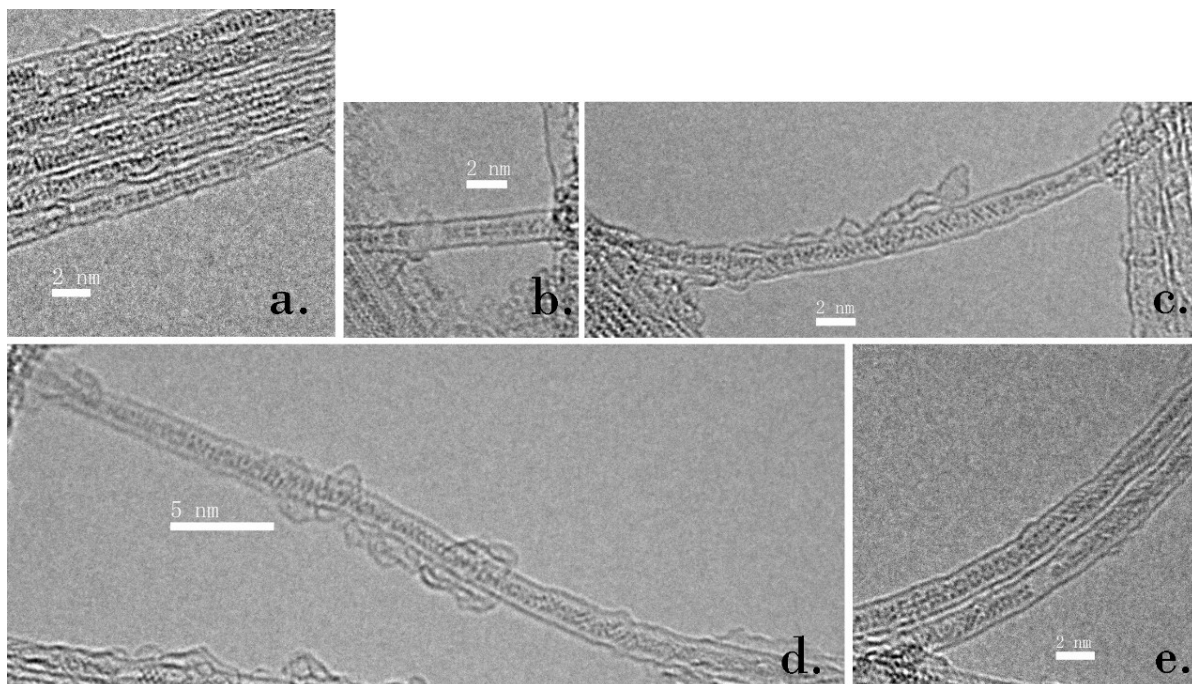
Supplementary Figure 9. HAADF images of ErCl₃@SWCNTs.



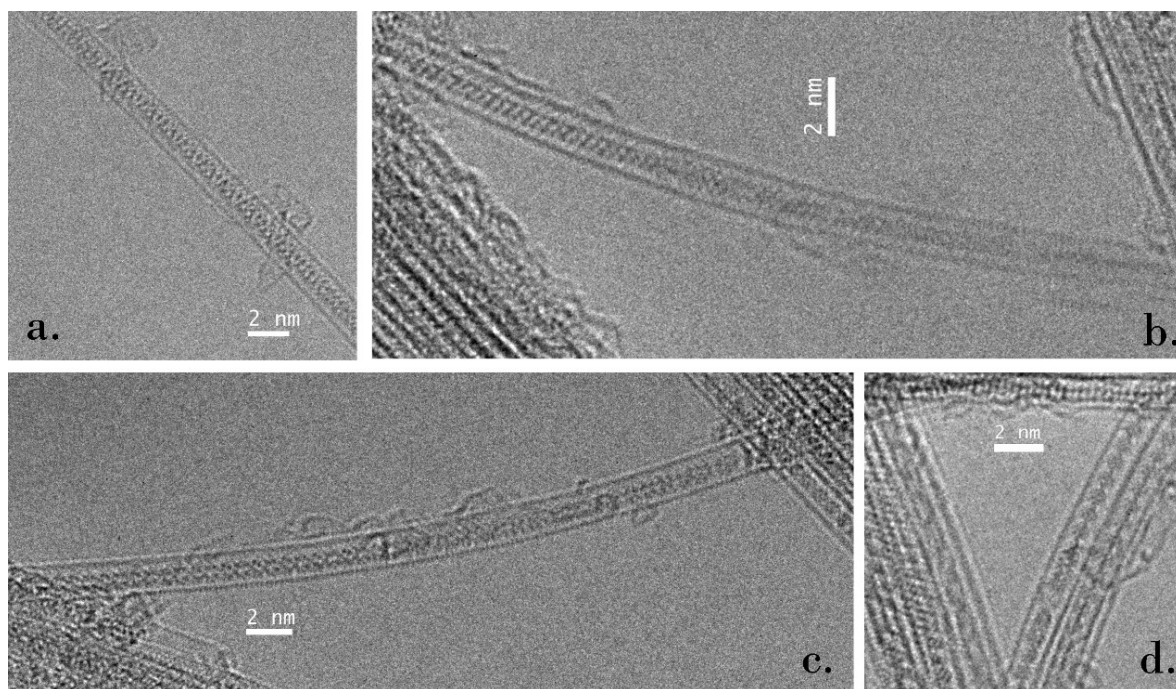
Supplementary Figure 10. TEM images of YCl₃@SWCNTs.



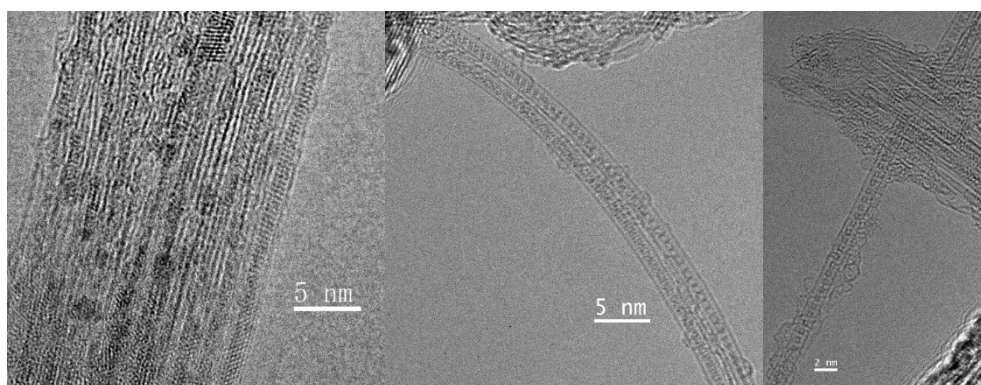
Supplementary Figure 11. TEM images of GdCl₃@SWCNTs.



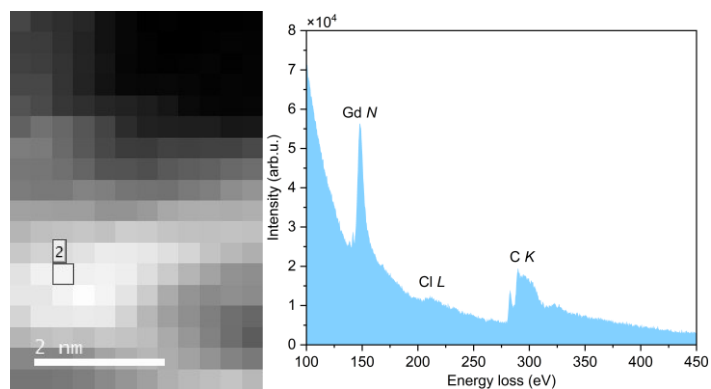
Supplementary Figure 12. TEM images of DyCl₃@SWCNTs.



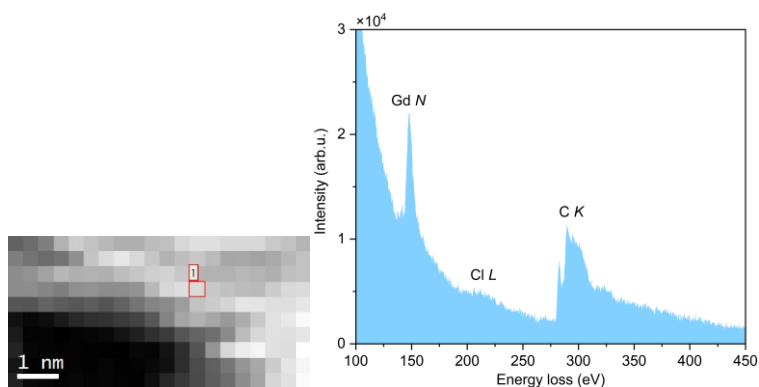
Supplementary Figure 13. HR-TEM images of DyCl₃@SWCNTs.



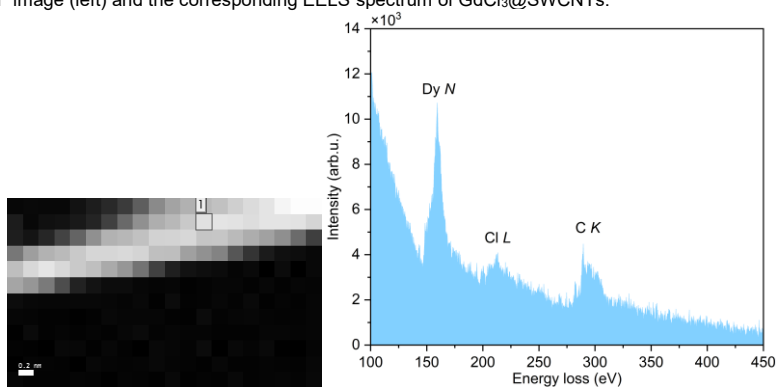
Supplementary Figure 14. HR-TEM images of ErCl₃@SWCNTs.



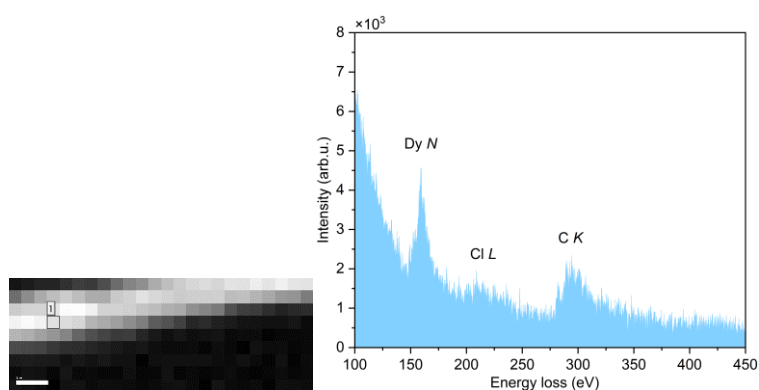
Supplementary Figure 15. HAADF image (left) and the corresponding EELS spectrum of GdCl₃@SWCNTs.



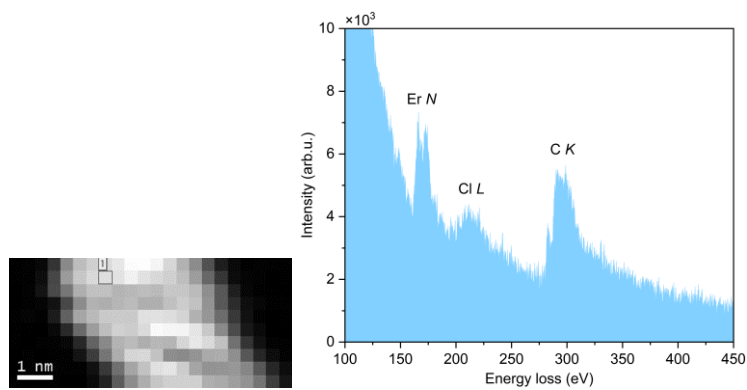
Supplementary Figure 16. HAADF image (left) and the corresponding EELS spectrum of GdCl₃@SWCNTs.



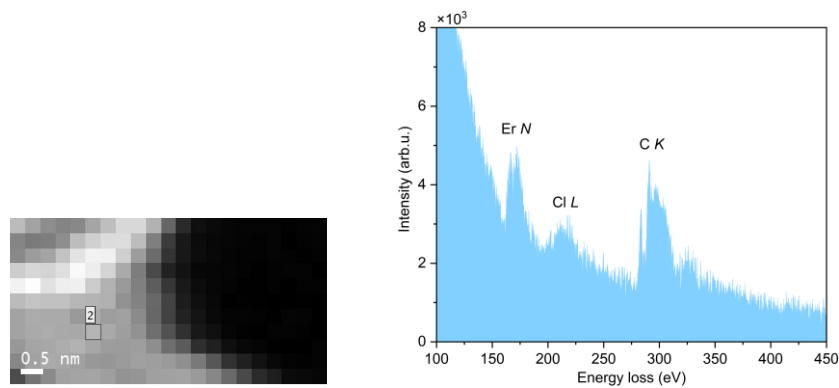
Supplementary Figure 17. HAADF image (left) and the corresponding EELS spectrum of DyCl₃@SWCNTs.



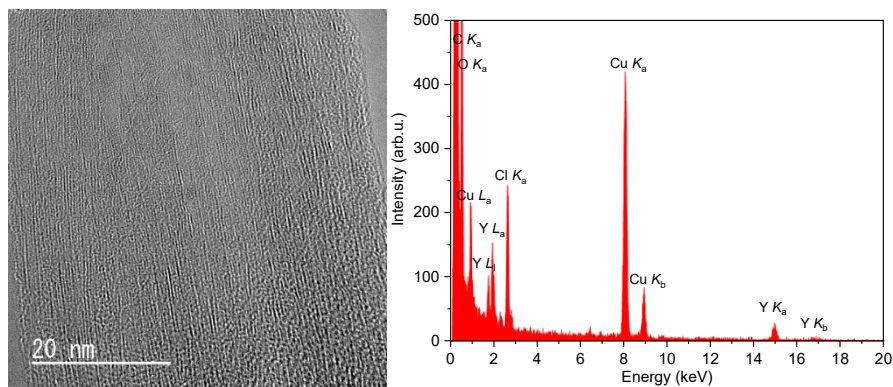
Supplementary Figure 18. HAADF image (left) and the corresponding EELS spectrum of DyCl₃@SWCNTs.



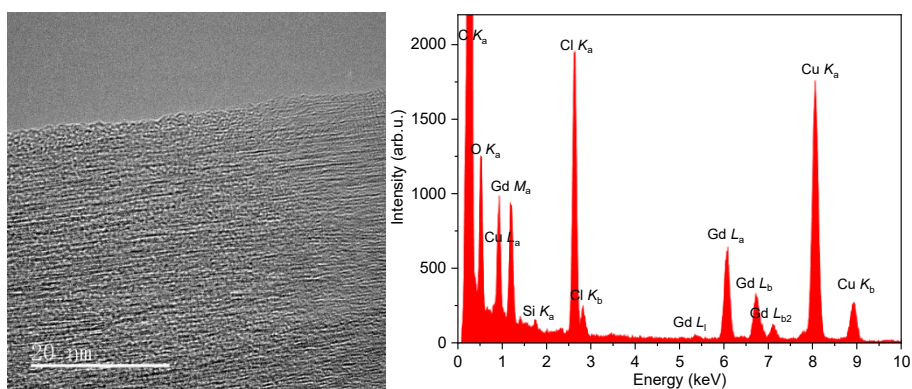
Supplementary Figure 19. HAADF image (left) and the corresponding EELS spectrum of $\text{ErCl}_3\text{@SWCNTs}$.



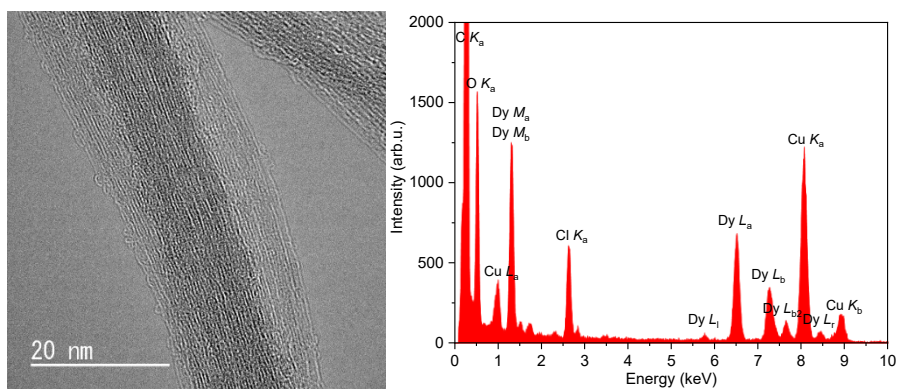
Supplementary Figure 20. HAADF image (left) and the corresponding EELS spectrum of $\text{ErCl}_3\text{@SWCNTs}$.



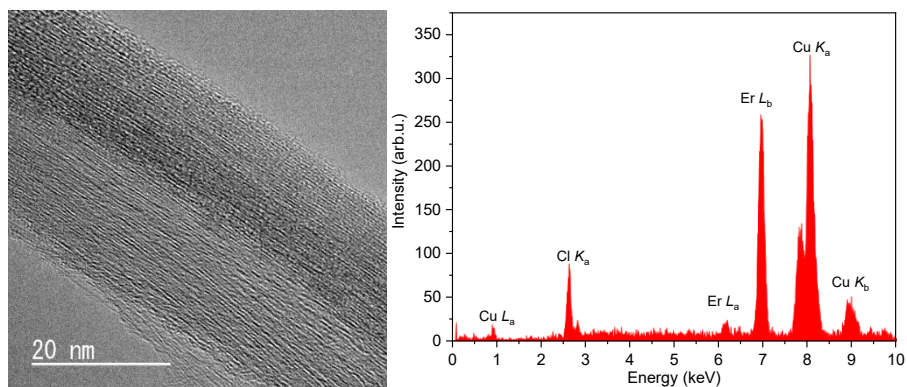
Supplementary Figure 21. TEM image (left) and corresponding EDX spectrum (right) of $\text{YCl}_3\text{@SWCNTs}$.



Supplementary Figure 22. TEM image (left) and corresponding EDX spectrum (right) of $\text{GdCl}_3\text{@SWCNTs}$.



Supplementary Figure 23. TEM image (left) and corresponding EDX spectrum (right) of DyCl₃@SWCNTs.



Supplementary Figure 24. TEM image (left) and corresponding EDX spectrum (right) of ErCl₃@SWCNTs.

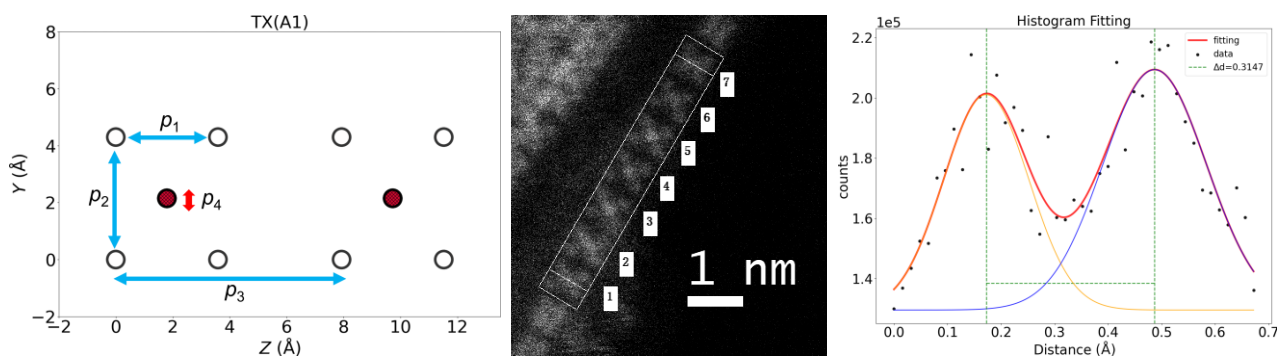
3 Guessing metal atom positions from the STEM-HADDF images

In order to obtain reasonable $\{\text{LnCl}_3\}_n$ nanowire geometries, it is necessary to have a preliminary and accurate guess of the location of the metal atoms. In this section, we analyzed STEM-HADDF data showing particular geometrical patterns. Here, two or more Gaussian functions were used for fitting the local signal counts in order to obtain information about the parameters (*i.e.*, p_1 , p_2 , p_3) used to describe the structural units. Here one case of the fitting results is given, more pictures plotted by the Gaussian fitting results can be found in the Supplementary Data 4 (STEMPatternMeasure). We measured different local structural units and averaged them as a base value for the conjecture.

For ease of discussion and without loss of generality, we define the plane in which the STEM image is profiled as the YZ plane and specify the direction of the nanowire extension as the Z axis. Through the above steps, only the information in the YZ projection plane can be obtained. The “depth” in the X-direction needs to be guessed (p_4). In some cases, we allow some metal atoms to rotate in the XY or XZ plane, so another parameter has to be given (p_5). The positions of the metal atoms are indicated by circles in the figure, their filled colors (blue-white-red) indicate their X-coordinates (low-high), and a mesh texture is used to emphasize the fact that if the two atoms projected in the YZ-plane are exactly coincident.

3.1 Type “X” (TX)

In the TX structure, we guess that its structural unit should have 6 metal atoms, forming an octahedral structure, thus on the YZ projection plane, it appears as an “X” pattern



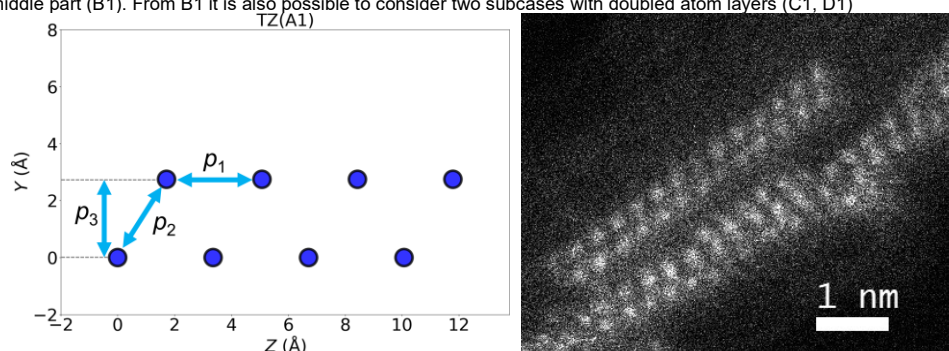
Supplementary Figure 25. The 2D projection of the TX model (left). Measurements of individual units to obtain parameters (middle). Intensity information is fitted using a double Gaussian function (right).

Supplementary Table 3. TX model parameters obtained by HADDF image fitting.

TX (Å)	p_1	p_2	p_3
	3.448	4.189	7.457
	3.147	4.428	8.057
	3.516	3.849	8.431
	3.949	4.465	7.768
	3.341	4.937	7.859
	3.812	4.285	8.021
	3.428	4.871	
	3.604	4.043	
	4.058	4.955	
		4.231	
		3.733	
		3.601	
avg.	3.589	4.299	7.932
std.	0.297	0.457	0.326

3.2 Type “Zigzag” (TZ)

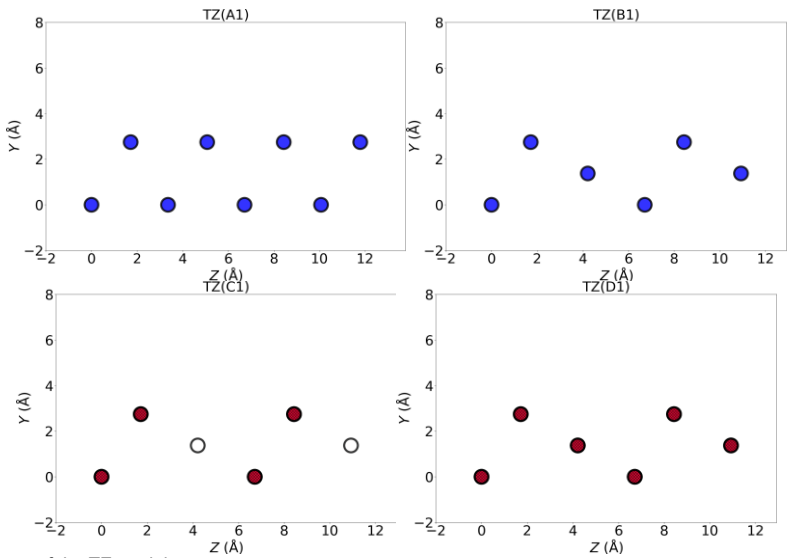
This is probably the simplest structure with two lines (A1). We also considered possible derived structures, such as doubling the supercell of A1, but placing only one metal atom in the middle part (B1). From B1 it is also possible to consider two subcases with doubled atom layers (C1, D1)



Supplementary Figure 26. The 2D projection of the TZ model (left). Measurements of individual units to obtain parameters (right).

Supplementary Table 4. TZ model parameters obtained by HADDF image fitting.

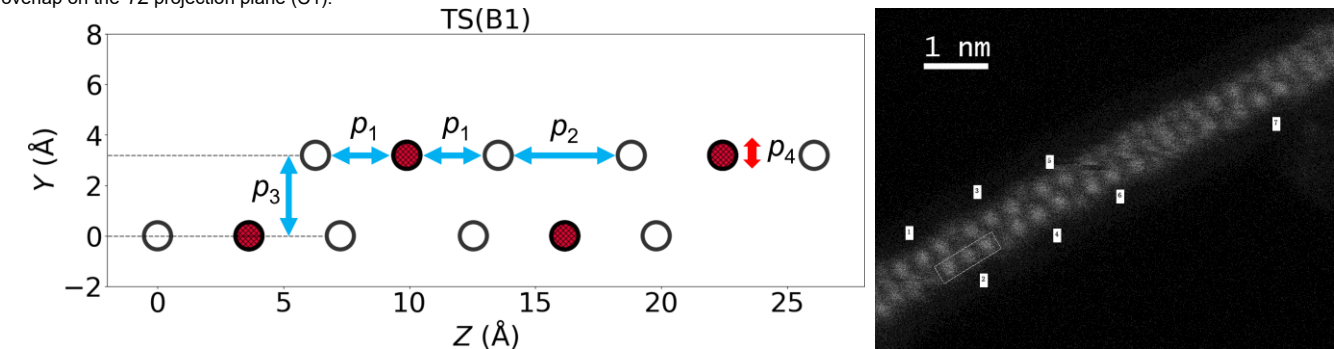
TZ (Å)	p_1	p_2	p_3
3.383	3.568	3.023	
3.386	3.490	2.951	
3.541	3.320	2.774	
3.211	3.966	2.426	
3.374	3.113	2.909	
3.487	2.556	2.477	
3.458	2.667	2.669	
3.522			
3.254			
3.541			
3.121			
3.309			
3.437			
2.963			
avg.	3.356	3.240	2.747
std.	0.170	0.503	0.234



Supplementary Figure 27. Subcases of the TZ model.

3.3 Type “Staggered” (TS)

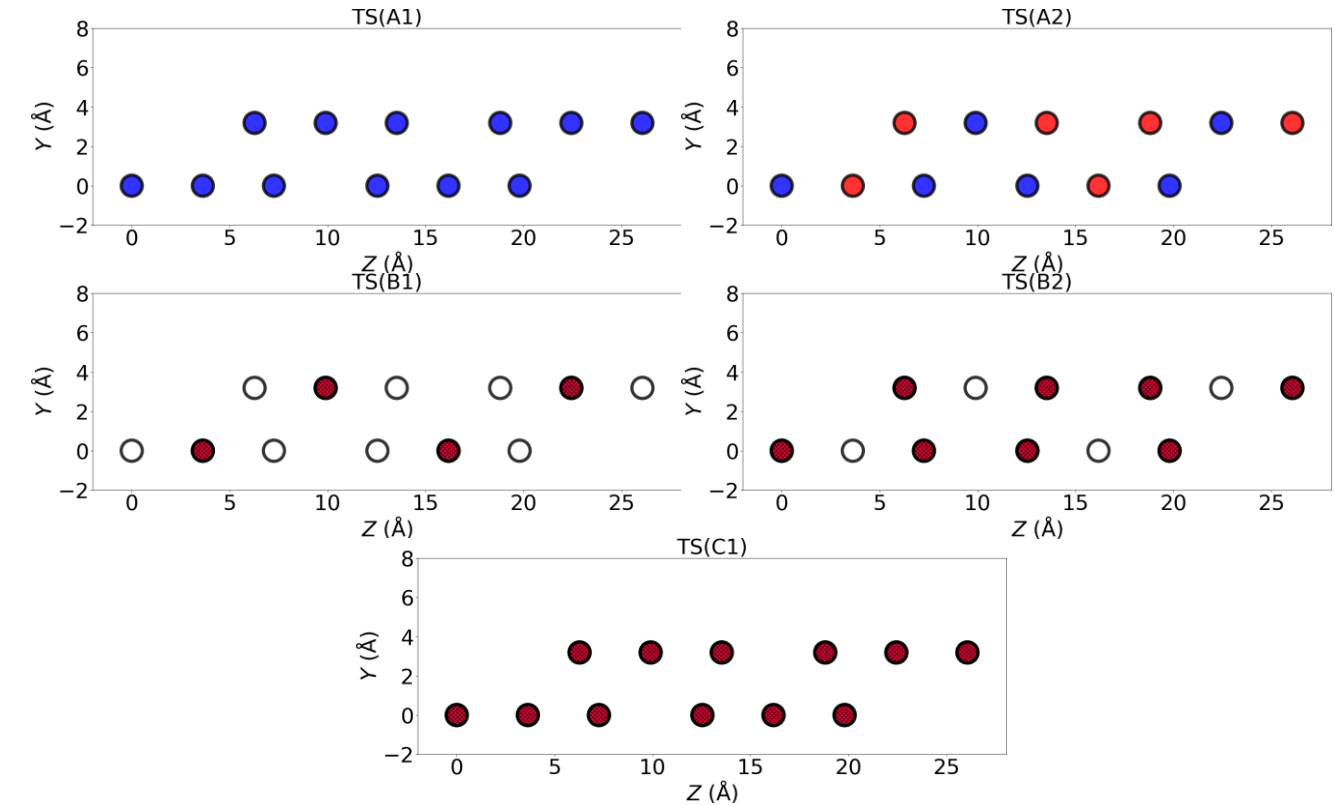
The TS structure is jagged. We guess that it may have only one layer of metal atoms in the same X plane (A1), or undulating in the X direction (B1). It is also possible that there are two layers of metal atoms (B1, B2) at certain positions. Of course, it is also possible that there are two layers of metal atoms that perfectly overlap on the YZ projection plane (C1).



Supplementary Figure 28. The 2D projection of the TS model (left). Measurements of individual units to obtain parameters (right).

Supplementary Table 5. TS model parameters obtained by HADDF image fitting.

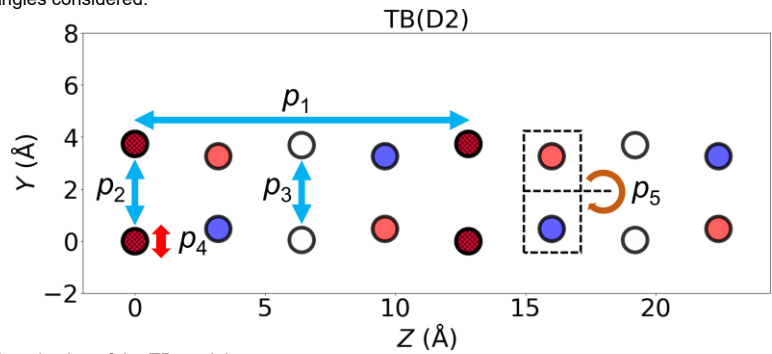
TS (Å)	p_1	p_2	p_3
3.628	5.282	3.192	
3.407	5.517	3.237	
3.567	5.511	3.092	
3.604	5.571	3.222	
3.536			
3.416			
3.279			
3.295			
3.694			
3.868			
4.007			
3.660			
avg.	3.580	5.470	3.186
std.	0.217	0.129	0.065



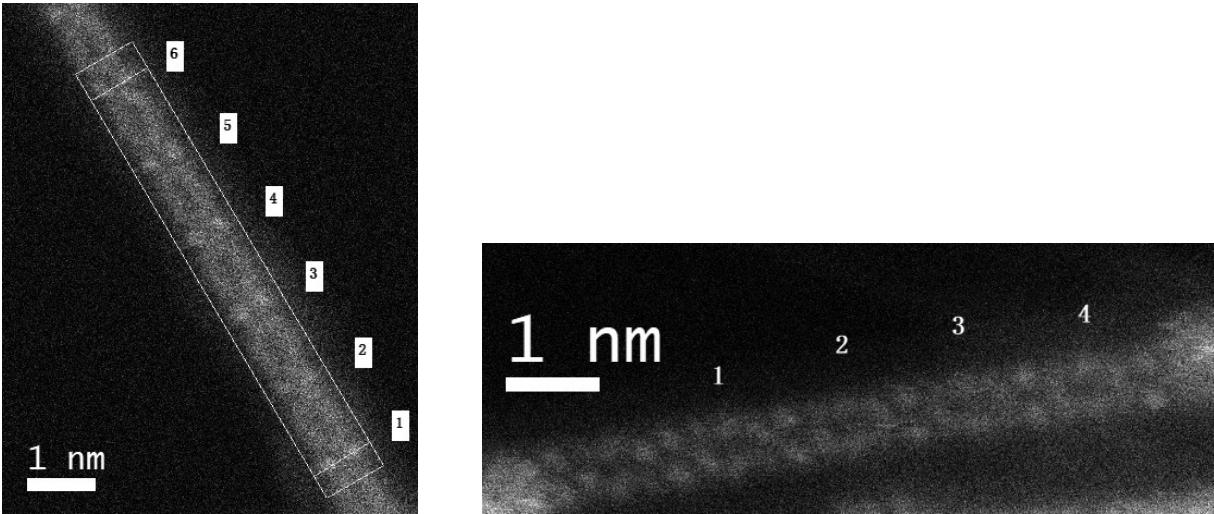
Supplementary Figure 29. Subcases of the TS model.

3.4 Type “Bamboo” (TB)

This structure shows a pattern similar to bamboo nodes. Two discrete bright spots with high contrast can be clearly observed at the node parts. The contrast of the middle part of the structural unit is less than the nodes, making it difficult to observe the discrete points. Moreover, the width between the middle parts of the structural units is slightly shorter than the width at the node parts. Therefore, we propose the following conjectural structure. Where A1 and B1 are monolayer structures and their middle parts have 4, 6 metal atoms, respectively. C1, D1, D2 are similar to the above structures except that two layers of atoms are considered at the node parts. For D2, we allow the pair of metal atoms close to the node to be rotatable in the XY plane, the angles (p_5) considered are (10°, 20°, 30°, 40°, 50°, 60°, 70°). E1, F1, and F2 are considered for the possibility of a double layer of metal in the middle parts, with 6 and 8 metal atoms, respectively. F2 is allowed to have similar rotation possibilities as D2, with the same angles considered.



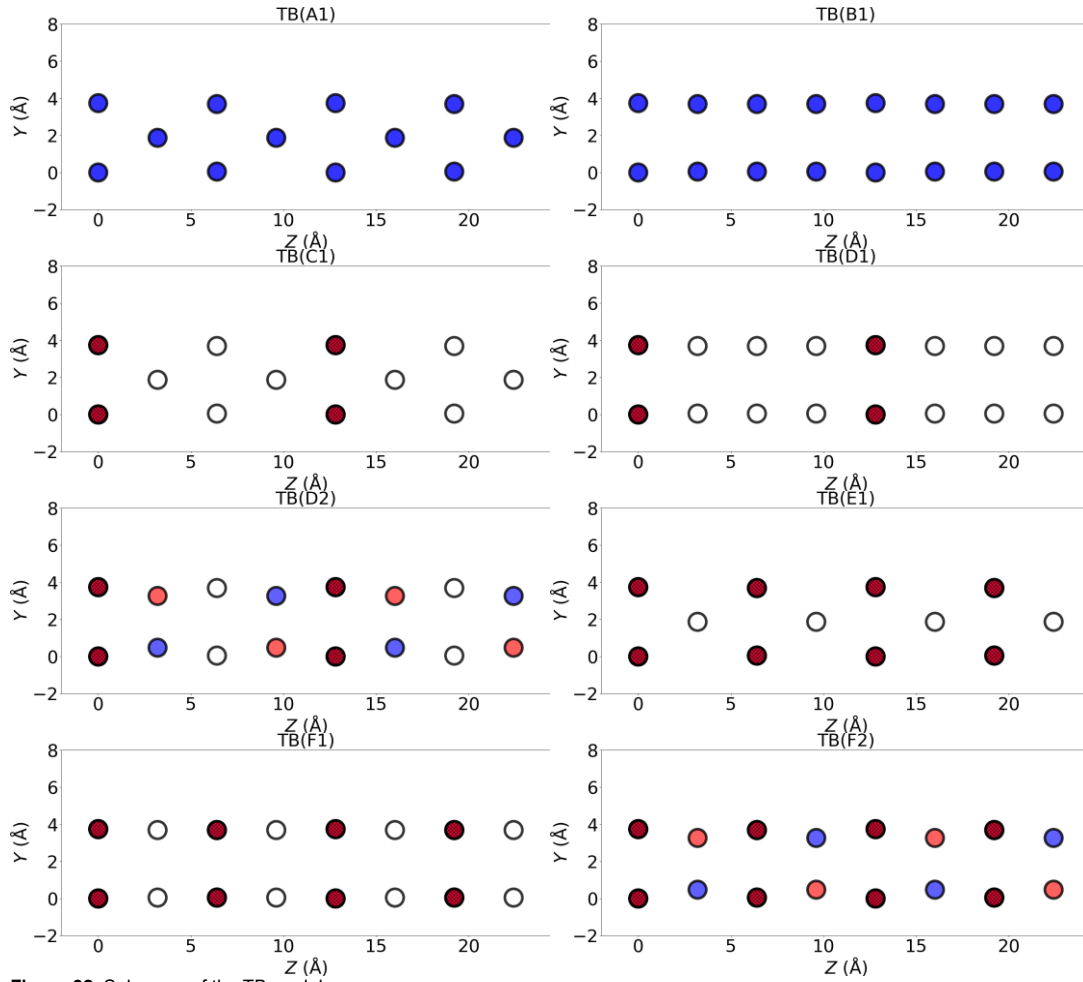
Supplementary Figure 30. The 2D projection of the TB model.



Supplementary Figure 31. Measurements of individual units to obtain parameters.

Supplementary Table 6. TB model parameters obtained by HADDF image fitting.

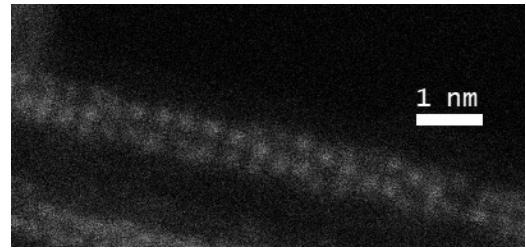
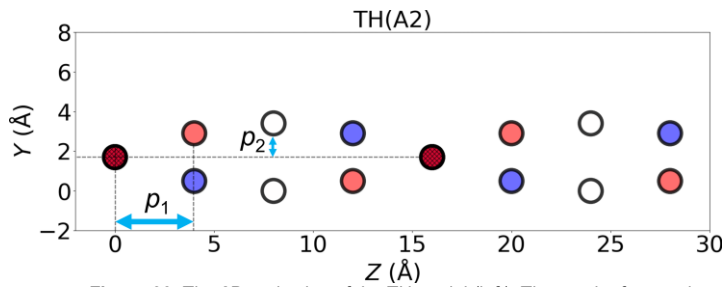
TB (Å)	p_1	p_2	p_3
12.406	3.824	3.876	
13.007	3.532	3.865	
12.693	3.502	3.711	
12.487	3.557	3.477	
12.454	3.529	3.413	
12.788	3.637	3.713	
12.885	3.638	3.710	
13.160	4.344	3.856	
13.097	3.966	3.876	
13.449	3.861	3.789	
12.214	3.691	3.509	
12.854	3.784	3.652	
12.534	3.816	3.482	
13.071	3.707	3.169	
12.880	3.618	3.516	
12.918	3.613	3.597	
		3.885	
avg.	12.806	3.736	3.638
std.	0.325	0.209	0.201



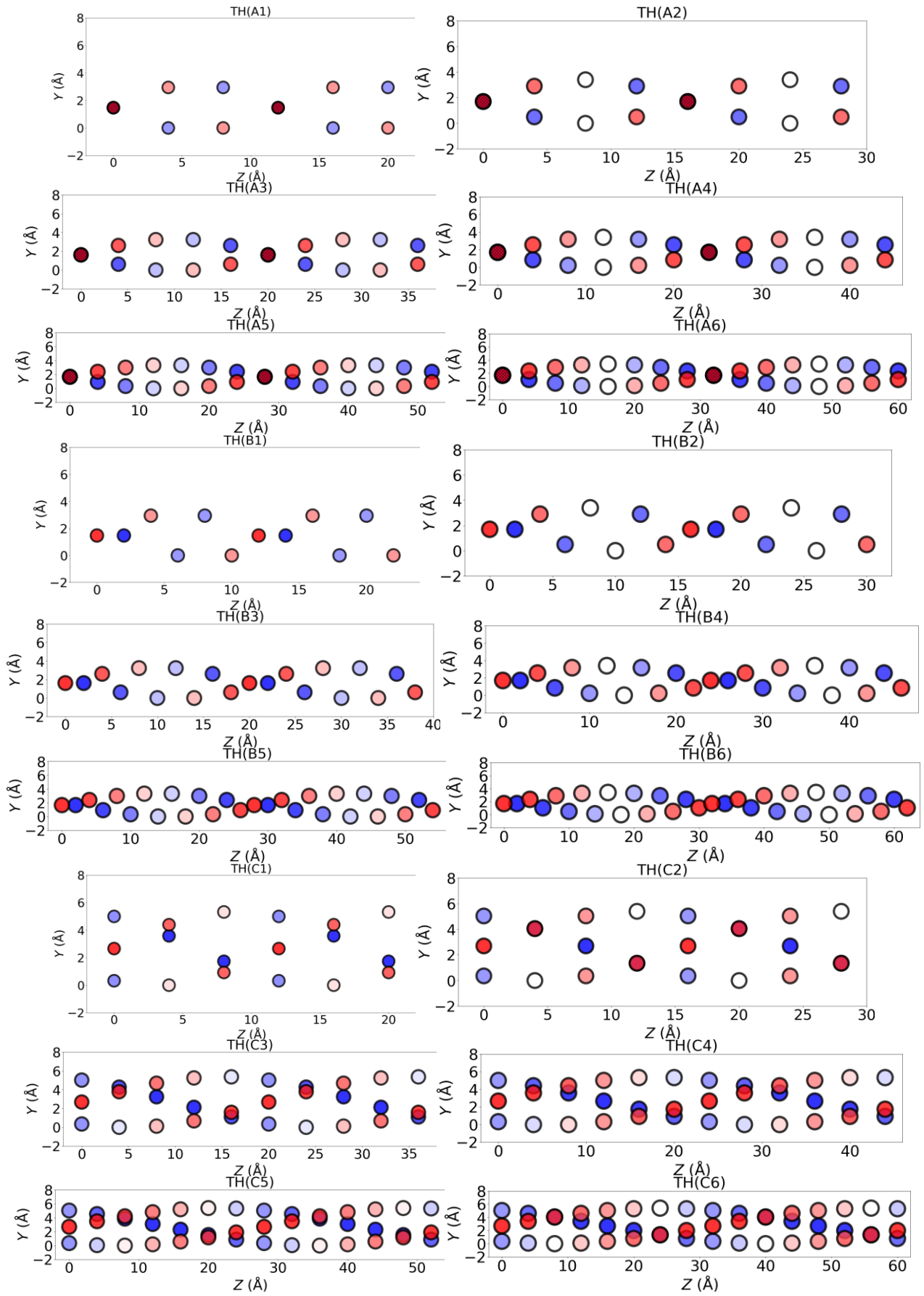
Supplementary Figure 32. Subcases of the TB model.

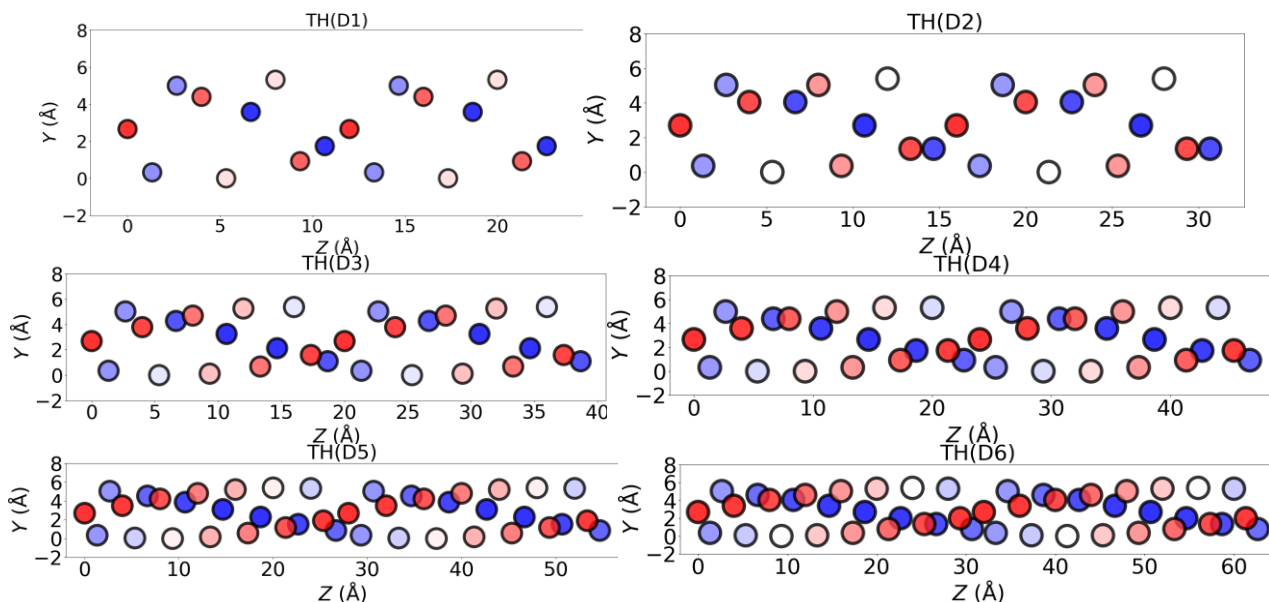
3.5 Type “helix” (TH)

The helix structure is also an uncommon case, but this is very natural and interesting so we have considered it as well. The p_1 parameter is the spacing in the Z-direction between two neighboring groups, and the p_2 parameter is the radius between the individual atoms from the central axis. Here, $p_1 = 4.0$ Å, $p_2 = 1.7$ Å for subcases A, B (double helix) and 2.7 Å for subcases C, D (triple helix). For An , each structural unit contains $(n+2)$ groups, each of which is rotated $360^\circ/(n+2)$ along the Z-axis relative to the previous group. For example, A1 contains three groups per structural unit, and each group is rotated along the Z-axis by 120° . The same rotational scheme applied for B, C, D subcases as well. The atoms within the same group in cases A and C, all have the same Z-coordinate. However, the two atoms within the same group in case B have different additional displacements in the Z direction of $(0, p_1/2)$, respectively. The D subcases are similar but with additional displacements of $(0, p_1/3, 2 p_1/3)$.



Supplementary Figure 33. The 2D projection of the TH model (left). The used reference image (right).

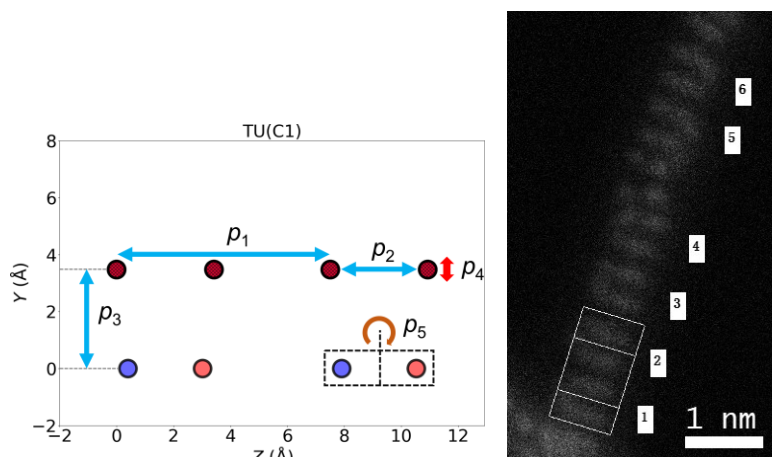




Supplementary Figure 34. Subcases of the TH model.

3.6 Type “U” (TU)

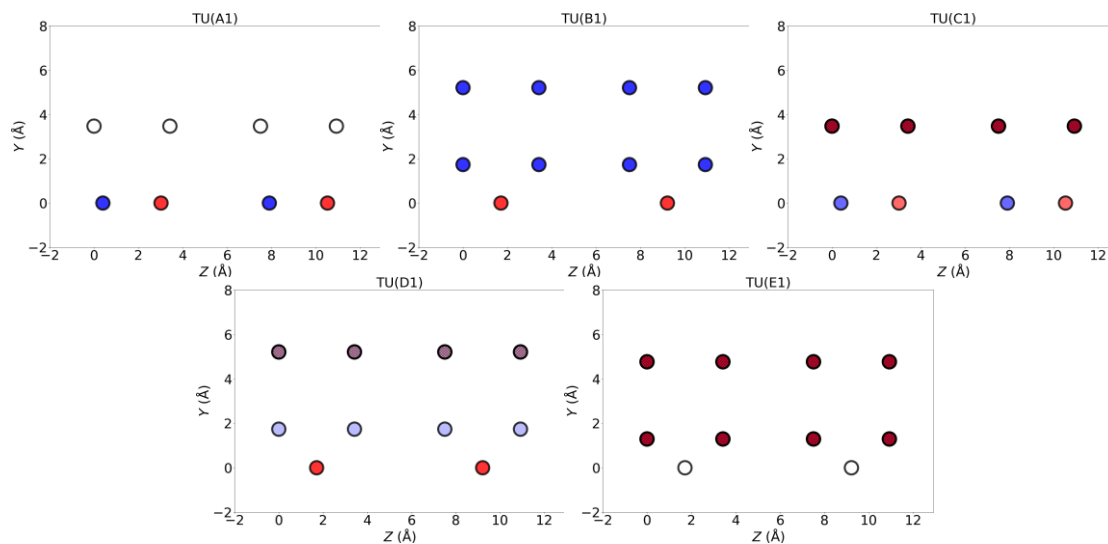
This structure looks similar to the shape of a cup from the HAADF image. A1 and B1 have only one layer of metal atoms and at the bottom of the cup they have two and three atoms, respectively. The two atoms at the bottom of A1 are allowed to rotate in the XZ plane and the chosen angles (p_5) are (10°, 20°, 30°, 40°, 50°, 60°). One atom at the bottom of B1 is allowed to rotate in the YZ plane and the chosen angles are (30°, 40°, 50°, 60°, 70°, 80°, 90°). C1 is similar to A1 and D1 is similar to B1 but with two layers of atoms placed at the top. E1 has two layers of atoms placed at the top and five atoms at the bottom.



Supplementary Figure 35. The 2D projection of the TU model (left). Measurements of individual units to obtain parameters (right).

Supplementary Table 7. TU model parameters obtained by HADDF image fitting.

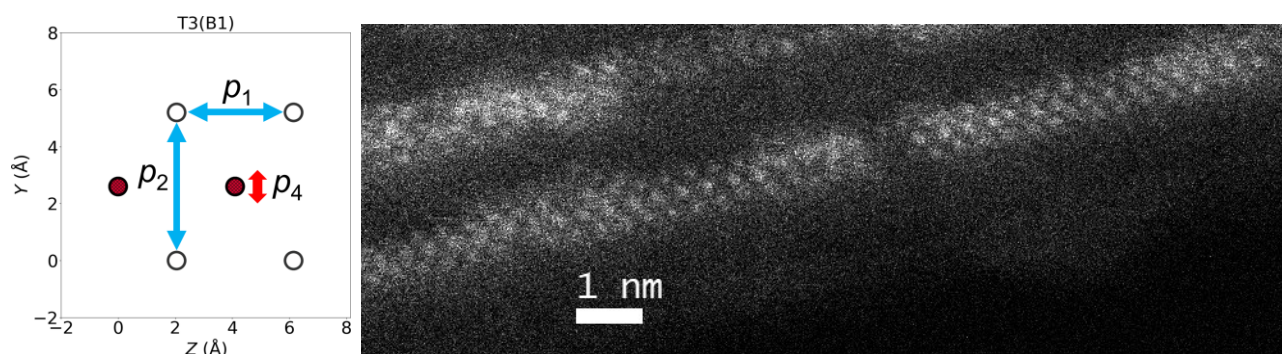
TU (Å)	p_1	p_2	p_3
	7.088	3.576	3.245
	8.077	3.306	3.317
	7.207	3.257	3.551
	7.662	3.847	3.588
		3.236	3.670
		3.256	3.601
		3.540	3.457
		3.364	3.300
			3.542
avg.	7.509	3.423	3.474
std.	0.453	0.215	0.152



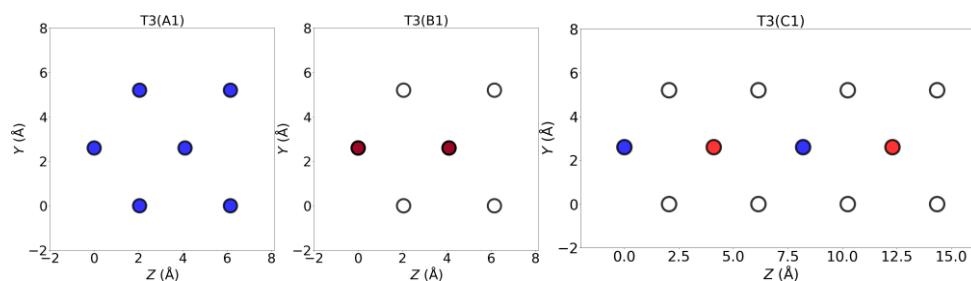
Supplementary Figure 36. Subcases of the TU model.

3.7 Type “three lines” (T3)

The structure with three lines is not common, the values ($p_1 = 4.1$ Å, $p_2 = 5.2$ Å) are roughly estimated based on the HADDF image. A1 has a single layer of metal atoms, B1 has two layers of metal in the middle part and a single layer on both sides. C1 also has a single layer but there is an undulation of atoms in the middle.



Supplementary Figure 37. The 2D projection of the T3 model (left). The used reference image (right).



Supplementary Figure 38. Subcases of the T3 model.

3.8 A short Summation of guessing configurations

Since we cannot guarantee that there is no gap due to systematic errors between the geometric structure information drawn from the STEM images and the subsequent optimization method, we need to scale the parameters (p_1, p_2, p_3) by a factor in the list (0.8, 0.9, 1.0, 1.1, 1.2, 1.3, 1.4). In some cases, the X-depth information is involved (p_4), and here we use the parameter list (2.0, 3.0, 4.0, 5.0, 6.0, 7.0). The information of rotation angle (p_5) has been mentioned above. We summarize the number of configurations generated with the above conjectures in the following table.

Supplementary Table 8. The number of configurations for the models and their subcases.

Type	Subcase	Config.	#(Ln)	Type	Subcase	Config.	#(Ln)
TX	A1	2058	6	TH	A1	49	6
TS	A1	343	6	TH	A2	49	8
TS	A2	2058	6	TH	A3	49	10
TS	B1	2058	8	TH	A4	49	12
TS	B2	2058	10	TH	A5	49	14
TS	C1	2058	12	TH	A6	49	16
TB	A1	343	6	TH	B1	49	6
TB	B1	343	8	TH	B2	49	8
TB	C1	2058	8	TH	B3	49	10
TB	D1	2058	10	TH	B4	49	12
TB	E1	2058	10	TH	B5	49	14
TB	F1	2058	10	TH	B6	49	16
TB	D2	14406	12	TH	C1	49	9
TB	F2	14406	12	TH	C2	49	12
TU	A1	2058	4	TH	C3	49	15
TU	B1	2401	5	TH	C4	49	18
TU	C1	12348	6	TH	C5	49	21
TU	D1	14406	7	TH	C6	49	24
TU	E1	2058	9	TH	D1	49	9
TZ	A1	343	4	TH	D2	49	12
TZ	B1	343	3	TH	D3	49	15
TZ	C1	2058	5	TH	D4	49	18
TZ	D1	2058	6	TH	D5	49	21
T3	A1	49	3	TH	D6	49	24
T3	B1	294	4				
T3	C1	294	6				
Total configurations:							88249
Number of MD copies:							8
Number of tasks:							705992

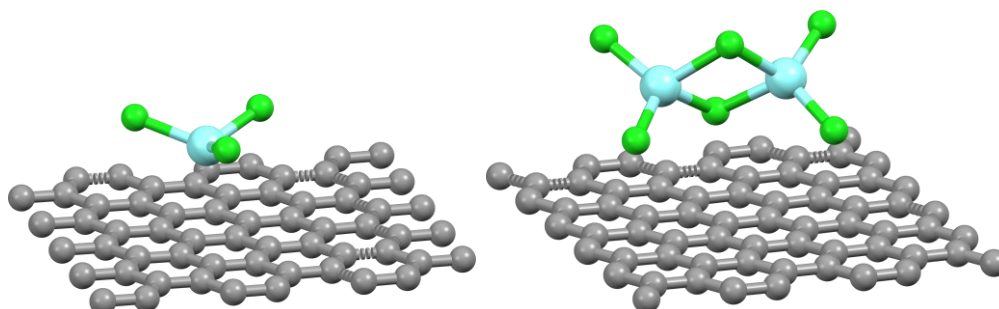
4 Exploring the structure

Supplementary Table 9. Unit cell parameters and average bond lengths were compared between DFT-optimized structures and experimental measurements.

	YCl₃		LaCl₃		GdCl₃		DyCl₃		TmCl₃	
	Exp.	DFT	Exp.	DFT	Exp.	DFT	Exp.	DFT	Exp.	DFT
a (Å)	6.92	6.86	7.48	7.45	7.36	7.32	3.82	3.78	6.44	6.42
b (Å)	11.94	11.81	7.48	7.45	7.36	7.32	11.82	11.85	6.44	6.42
c (Å)	6.44	6.39	4.37	4.35	4.11	4.07	8.51	8.46	18.02	17.89
α (°)	90.0	90.0	90.0	90.0	90.0	90.0	90.0	90.0	90.0	90.0
β (°)	111.0	111.1	90.0	90.0	90.0	90.0	90.0	90.0	90.0	90.0
γ (°)	90.0	90.0	120.0	120.0	120.0	120.0	90.0	90.0	120.0	120.0
V (Å³)	496.8	482.6	211.9	209.0	192.7	189.0	383.6	378.1	646.7	639.4
dev. V	-2.85%		-1.37%		-1.96%		-1.41%		-1.11%	
avg.(Ln-Cl) (Å)	2.637	2.642	2.951	2.943	2.851	2.839	2.785	2.771	2.608	2.604
dev. avg.(Ln-Cl)	0.20%		-0.28%		-0.42%		-0.48%		-0.12%	
	Graphite									
	Exp.	DFT								
a (Å)	2.46	2.47								
b (Å)	2.46	2.47								
c (Å)	6.71	6.69								
α (°)	90.0	90.0								
β (°)	90.0	90.0								
γ (°)	120.0	120.0								
V (Å³)	35.25	35.21								
dev V	-0.12%									
avg.(C-C) (Å)	1.422	1.423								
dev. avg.(C-C)	0.13%									

Supplementary Table 10. DFT-optimized unit cell parameters, bond lengths, and relative energies were obtained for yttrium-replaced LnCl₃ crystals of various space groups.

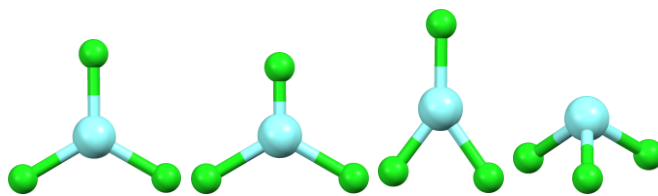
Space Group	C2/m	P6₃/m	P4₂/mmn	Cmcm	R3̄c
Space Group Number	12	176	136	63	167
Reference compound	YCl ₃	GdCl ₃	TbCl ₃	DyCl ₃	TmCl ₃
The number of YCl ₃ (Z)	4	2	4	4	6
V _{YCl₃} (Å ³)	482.6	187.1	461.8	379.9	660.3
V _{LnCl₃Exp.} (Å ³)	496.8	192.7	486.0	383.6	646.7
dev. V	-2.85%	-2.91%	-4.98%	-0.94%	2.11%
avg.(Y-Cl) (Å)	2.64	2.82	2.64	2.77	2.64
avg.(Ln-Cl) Exp. (Å)	2.64	2.85	2.71	2.78	2.61
dev. avg.(Ln-Cl)	0.20%	-0.91%	-2.56%	-0.36%	1.23%
E (eV)	-87.5304	-43.8032	-87.4063	-87.7883	-131.1289
E / Z (eV)	-21.8826	-21.9016	-21.8516	-21.9471	-21.8548
Ē (eV)	-2.7826	-2.8016	-2.7516	-2.8471	-2.7548



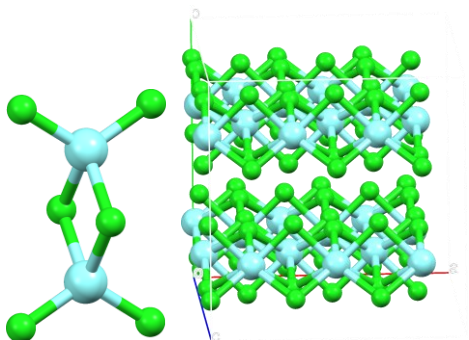
Supplementary Figure 39. Ball-and-stick models of the YCl₃@Graphene (left) and Y₂Cl₆@Graphene(right) are shown. The colors of the atoms indicate yttrium (blue), chlorine (green), and carbon (gray).

Supplementary Table 11. DFT energy of each system after DFT optimization.

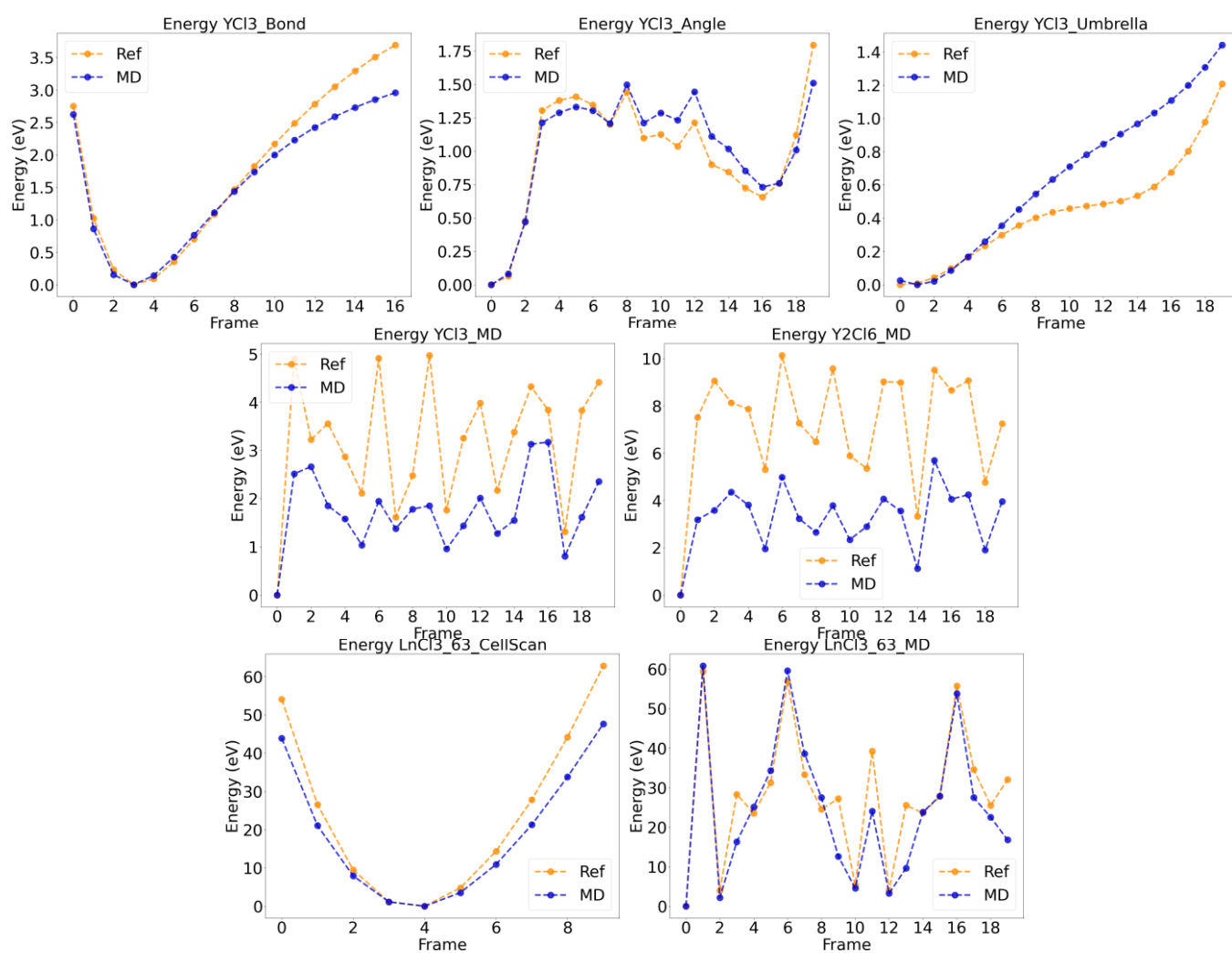
	YCl₃	Graphene	
E (eV)	-19.1000	-559.3455	
	Y ₂ Cl ₆	YCl ₃ @Graphene	Y ₂ Cl ₆ @Graphene
E (eV)	-40.3432	-579.4670	-600.2690
ΔE (eV)		-1.0214	-0.5803



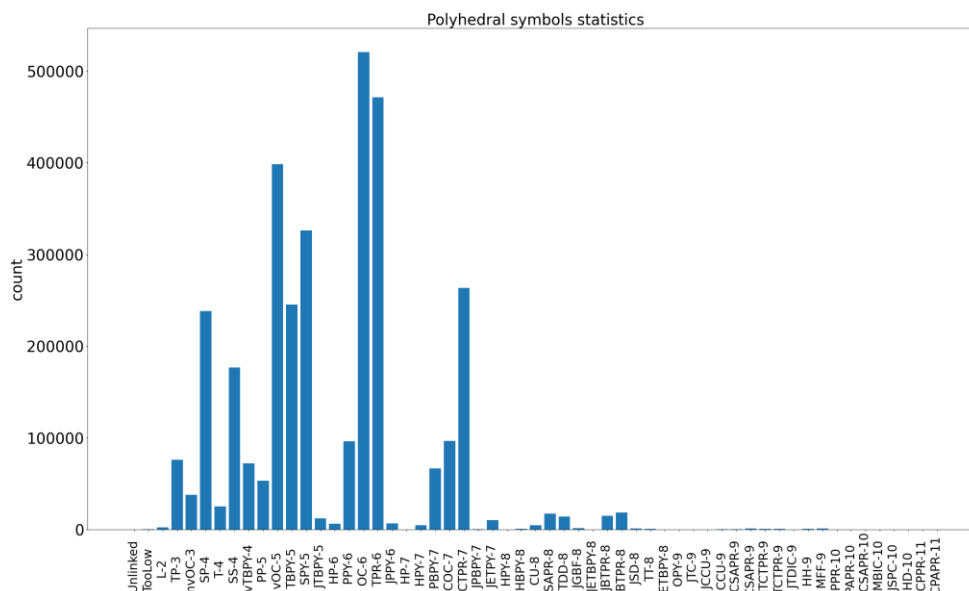
Supplementary Figure 40. The ball-and-stick models of the YCl_3 molecule. The colors of the atoms indicate yttrium (cyan) and chlorine (green).



Supplementary Figure 41. The ball-and-stick models of the Y_2Cl_6 molecule and YCl_3 crystal. The colors of the atoms indicate yttrium (cyan) and chlorine (green).



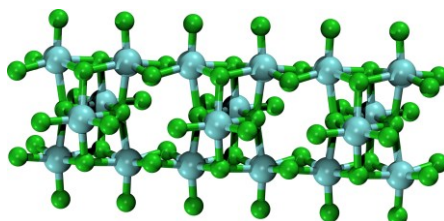
Supplementary Figure 42. The energies of the YCl_3 subsystems' frames after the optimization of the parameters. The yellow and the blue lines represent the reference and the ReaxFF-MD simulation results, respectively.



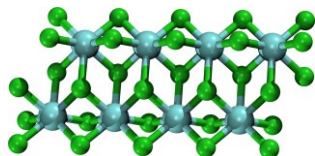
Supplementary Figure 43. Statistics of the number of polyhedra in all valid configurations after simulated annealing of chloride ions by ReaxFF-MD.

Supplementary Table 12 Polyhedron types and their abbreviations.

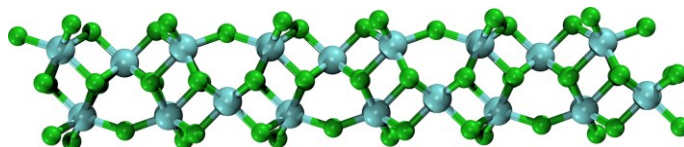
Symbol	Polygon	Symbol	Polygon	Symbol	Polygon	Symbol	Polygon
L-2	Linear	HP-7	Heptagon	EP-9	Enneagon	DP-10	Decagon
vT-2	Divacant tetrahedron	HPY-7	Hexagonal pyramid	OPY-9	Octagonal pyramid	EPY-10	Enneagonal pyramid
vOC-2	Tetravacant octahedron	PBPY-7	Pentagonal bipyramid	HBPY-9	Heptagonal bipyramid	OBPY-10	Octagonal bipyramid
TP-3	Trigonal	COC-7	Capped octahedron	JTC-9	Johnson triangular cupola	PPR-10	Pentagonal prism
vT-3	Vacant tetrahedron	CTPR-7	Capped trigonal prism	JCCU-9	Capped cube	PAPR-10	Pentagonal antiprism
fvOC-3	fac-Trivacant octahedron	JPBPY-7	Johnson pentagonal bipyramid	CCU-9	Spherical-relaxed capped cube	JBCCU-10	Bicapped cube
mvOC-3	mer-Trivacant octahedron	JETPY-7	Johnson elongated triangular pyramid	JCSAPR-9	Capped square antiprism	JBCSAPR-10	Bicapped square antiprism
SP-4	Square	OP-8	Octagon	CSAPR-9	Spherical capped square antiprism	JMBIC-10	Metabidiminshed icosahedron
T-4	Tetrahedron	HPY-8	Heptagonal pyramid	JTCTPR-9	Tricapped trigonal prism	JATDI-10	Augmented tridiminshed icosahedron
SS-4	Seesaw	HBPY-8	Hexagonal bipyramid	TCTPR-9	Spherical tricapped trigonal prism	JSPC-10	Sphenocorona
vTBPY-4	Axially vacant trigonal bipyramid	CU-8	Cube	JTDIC-9	Tridiminshed icosahedron	SDD-10	Staggered Dodecahedron
PP-5	Pentagon	SAPR-8	Square antiprism	HH-9	Hula-hoop	TD-10	Tetradecahedron
vOC-5	Vacant octahedron	TDD-8	Triangular dodecahedron	MFF-9	Muffin	HD-10	Hexadecahedron
TBPY-5	Trigonal bipyramid	JGBF-8	Johnson gyrobifastigium			HP-11	Hendecagon
SPY-5	Spherical square pyramid	JETBPY-8	Johnson elongated triangular bipyramid			DPY-11	Decagonal pyramid
JTBPY-5	Johnson trigonal bipyramid	JBTPR-8	Biaugmented trigonal prism			EBPY-11	Enneagonal bipyramid
HP-6	Hexagon	BTPR-8	Biaugmented trigonal prism			JCPPR-11	Capped pentagonal prism
PPY-6	Pentagonal pyramid	JSD-8	Snub diphenoid			JCPAPR-11	Capped pentagonal antiprism
OC-6	Octahedron	TT-8	Triakis tetrahedron			JAPPR-11	Augmented pentagonal prism
TPR-6	Trigonal prism	ETBPY-8	Elongated trigonal bipyramid			JASPC-11	Augmented sphenocorona



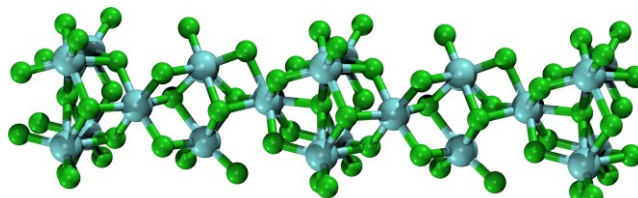
Supplementary Figure 44. Ball-and-stick models of the TX units are shown. The colours of the atoms indicate yttrium (blue) and chlorine (green).



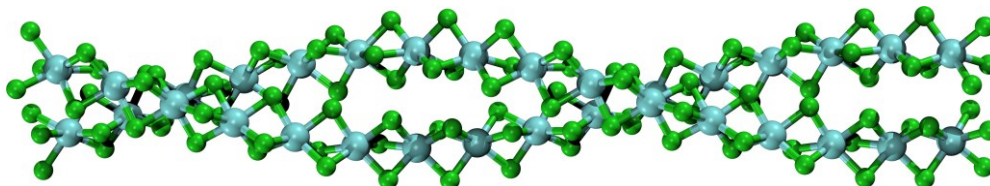
Supplementary Figure 45. Ball-and-stick models of the TZ units are shown. The colours of the atoms indicate yttrium (blue) and chlorine (green).



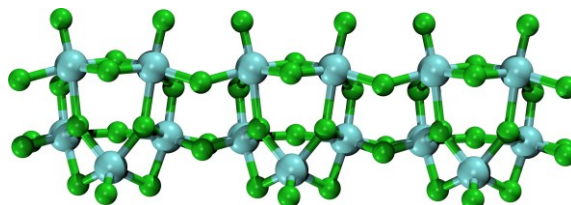
Supplementary Figure 46. Ball-and-stick models of the TS units are shown. The colours of the atoms indicate yttrium (blue) and chlorine (green).



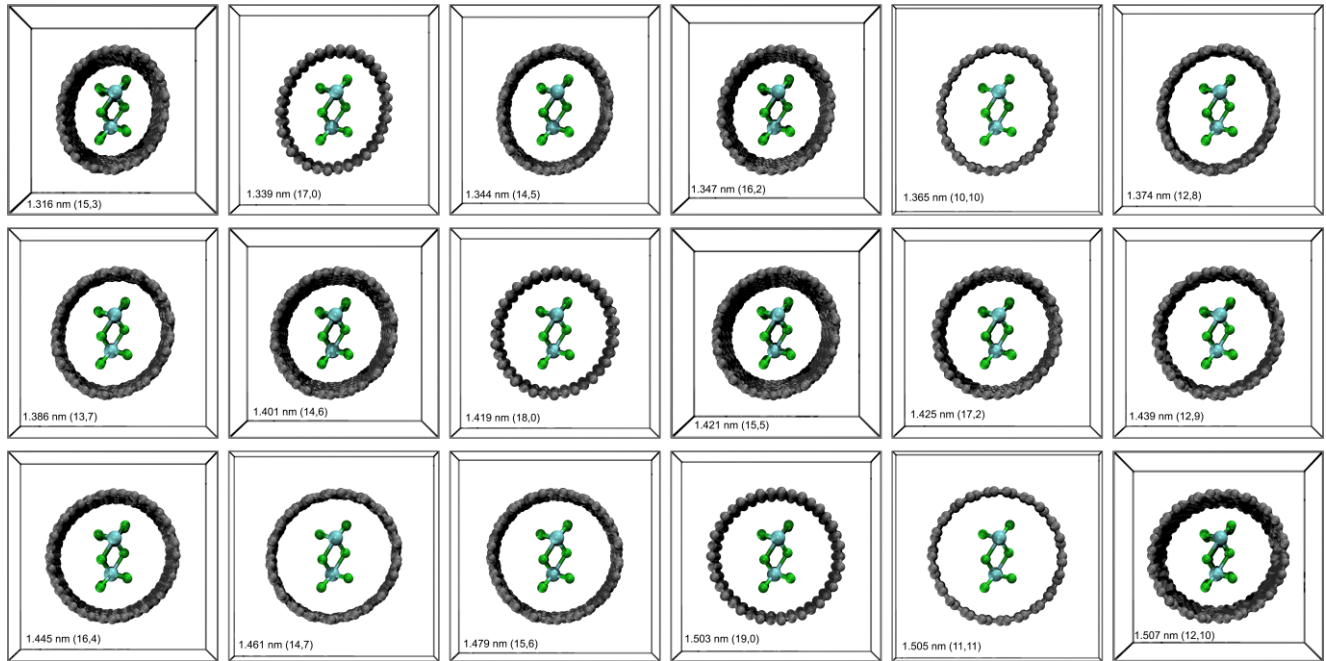
Supplementary Figure 47. Ball-and-stick models of the TB units are shown. The colours of the atoms indicate yttrium (blue) and chlorine (green).



Supplementary Figure 48. Ball-and-stick models of the TH units are shown. The colours of the atoms indicate yttrium (blue) and chlorine (green).



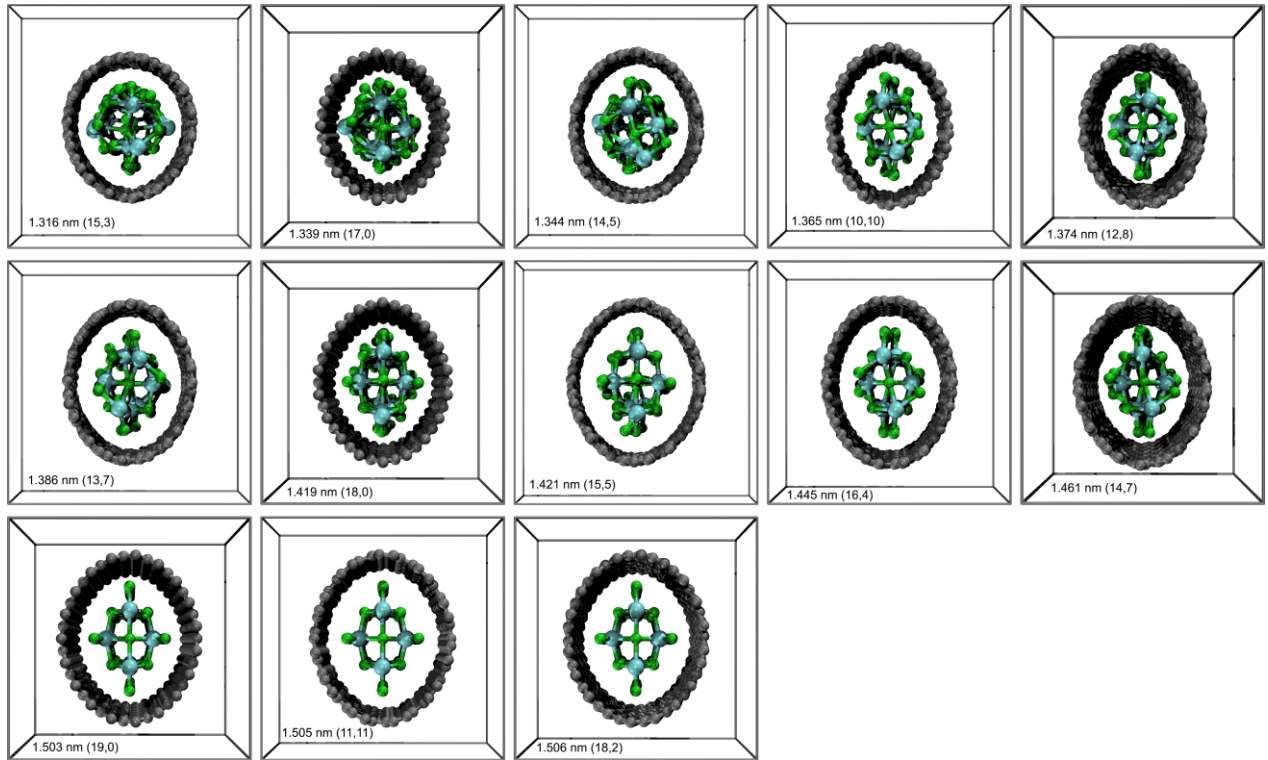
Supplementary Figure 49. Ball-and-stick models of the TU units are shown. The colours of the atoms indicate yttrium (blue) and chlorine (green).



Supplementary Figure 50. Axial views of TZ nanowire models encapsulated in SWCNTs of varying diameters.

Supplementary Table 13. Main structural parameters of TZ nanowires encapsulated in SWCNTs of varying diameters.

Hybrids	Pristine SWCNT's diameter (Å)	Pristine SWCNT's cell length (Å)	N_{swcnt}	Hybrid's cell length (Å)	Lattice mismatch	$\bar{E}_{\text{Hyb.}}$ (eV)	$\frac{m_{\text{YCl3}}}{m_{\text{YCl3@SWCNT}}}$
{Y ₂ Cl ₆ } ₁₃ @SWCNT(15,3)	13.16	23.762	2	47.403	-0.25%	-2.831	36.3%
{Y ₂ Cl ₆ } ₆ @SWCNT(17,0)	13.39	4.269	5	21.581	1.11%	-2.742	36.5%
{Y ₂ Cl ₆ } ₇ @SWCNT(14,5)	13.44	24.273	1	24.863	2.43%	-2.402	37.0%
{Y ₂ Cl ₆ } ₁₀ @SWCNT(16,2)	13.47	36.457	1	36.418	-0.11%	-2.836	35.8%
{Y ₂ Cl ₆ } ₂ @SWCNT(10,10)	13.65	2.463	3	7.333	-0.74%	-2.794	35.2%
{Y ₂ Cl ₆ } ₅ @SWCNT(12,8)	13.74	18.601	1	18.395	-1.11%	-2.686	34.9%
{Y ₂ Cl ₆ } ₇ @SWCNT(13,7)	13.86	24.994	1	25.233	0.96%	-2.749	35.6%
{Y ₂ Cl ₆ } ₁₀ @SWCNT(14,6)	14.01	37.920	1	37.153	-2.02%	-2.348	34.0%
{Y ₂ Cl ₆ } ₆ @SWCNT(18,0)	14.19	4.267	5	21.581	1.15%	-2.671	35.2%
{Y ₂ Cl ₆ } ₁₃ @SWCNT(15,5)	14.21	15.383	3	46.722	1.24%	-2.652	35.2%
{Y ₂ Cl ₆ } ₇ @SWCNT(17,2)	14.25	25.719	1	25.593	-0.49%	-2.729	34.3%
{Y ₂ Cl ₆ } ₇ @SWCNT(12,9)	14.39	25.947	1	25.710	-0.91%	-2.640	33.9%
{Y ₂ Cl ₆ } ₇ @SWCNT(16,4)	14.45	6.517	4	25.769	-1.14%	-2.565	33.7%
{Y ₂ Cl ₆ } ₃ @SWCNT(14,7)	14.61	11.288	1	11.102	-1.65%	-2.398	33.2%
{Y ₂ Cl ₆ } ₅ @SWCNT(15,6)	14.79	8.876	2	17.977	1.27%	-2.574	34.3%
{Y ₂ Cl ₆ } ₆ @SWCNT(19,0)	15.03	4.265	5	21.581	1.21%	-2.546	33.9%
{Y ₂ Cl ₆ } ₂ @SWCNT(11,11)	15.05	2.461	3	7.333	-0.67%	-2.593	33.0%
{Y ₂ Cl ₆ } ₁₁ @SWCNT(12,10)	15.07	40.674	1	40.361	-0.77%	-2.552	33.0%



Supplementary Figure 51. Axial views of TX nanowire models encapsulated in SWCNTs of varying diameters.

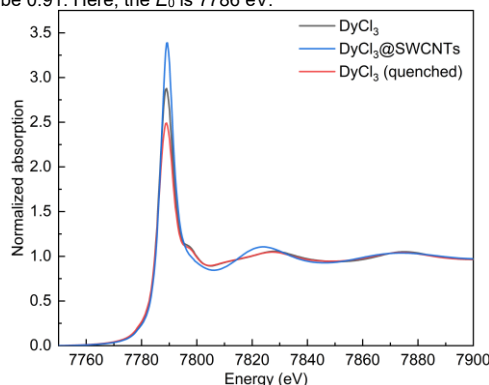
Supplementary Table 14. Main structural parameters of TX nanowires encapsulated in SWCNTs of varying diameters.

Hybrids	Pristine SWCNT's diameter (Å)	Pristine SWCNT's cell length (Å)	N_{swcnt}	Hybrid's cell length (Å)	Lattice mismatch	$\bar{E}_{\text{Hyb.}}$ (eV)	$\frac{m_{\text{YCl3}}}{m_{\text{YCl3@SWCNT}}}$
$\{\text{Y}_6\text{Cl}_{18}\}_3\text{@SWCNT}(15,3)$	13.16	23.762	1	23.924	0.68%	-2.018	44.1%
$\{\text{Y}_6\text{Cl}_{18}\}_6\text{@SWCNT}(17,0)$	13.39	4.269	11	47.558	1.29%	-1.898	43.9%
$\{\text{Y}_6\text{Cl}_{18}\}_3\text{@SWCNT}(14,5)$	13.44	24.273	1	24.177	-0.40%	-2.142	43.0%
$\{\text{Y}_6\text{Cl}_{18}\}_4\text{@SWCNT}(10,10)$	13.65	2.463	13	32.073	0.19%	-2.068	42.9%
$\{\text{Y}_6\text{Cl}_{18}\}_7\text{@SWCNT}(12,8)$	13.74	18.601	3	56.004	0.36%	-2.076	42.8%
$\{\text{Y}_6\text{Cl}_{18}\}_3\text{@SWCNT}(13,7)$	13.86	24.994	1	24.547	-1.79%	-1.967	41.6%
$\{\text{Y}_6\text{Cl}_{18}\}_6\text{@SWCNT}(18,0)$	14.19	4.267	11	47.558	1.32%	-2.088	42.5%
$\{\text{Y}_6\text{Cl}_{18}\}_2\text{@SWCNT}(15,5)$	14.21	15.383	1	15.722	2.21%	-1.907	42.9%
$\{\text{Y}_6\text{Cl}_{18}\}_4\text{@SWCNT}(16,4)$	14.45	6.517	5	32.356	-0.70%	-2.210	41.1%
$\{\text{Y}_6\text{Cl}_{18}\}_7\text{@SWCNT}(14,7)$	14.61	11.288	5	56.330	-0.19%	-2.252	41.1%
$\{\text{Y}_6\text{Cl}_{18}\}_6\text{@SWCNT}(19,0)$	15.03	4.265	11	47.558	1.38%	-2.111	41.2%
$\{\text{Y}_6\text{Cl}_{18}\}_4\text{@SWCNT}(11,11)$	15.05	2.461	13	32.073	0.25%	-2.227	40.6%
$\{\text{Y}_6\text{Cl}_{18}\}_5\text{@SWCNT}(18,2)$	15.06	40.677	1	40.428	-0.61%	-2.185	40.1%

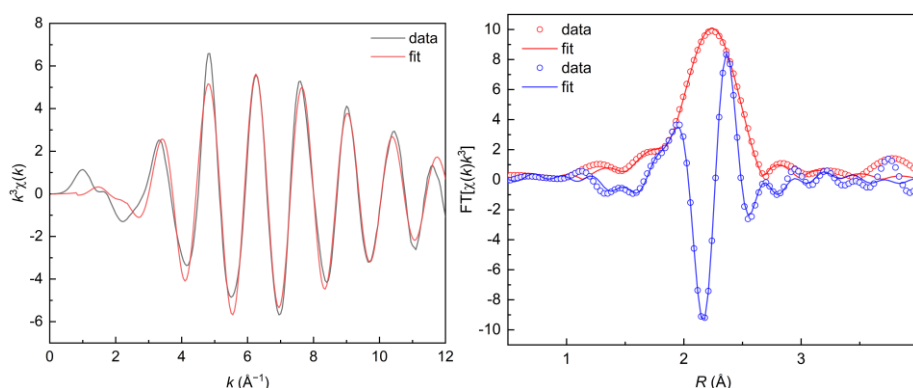
5 XAFS

5.1 DyCl₃

In the DyCl₃ case, the extracted k^3 -weighted extend X-ray absorption fine structure (EXAFS) oscillations over $k = 3\text{--}12 \text{ \AA}^{-1}$ after the smoothing background subtracted the XAFS spectra were Fourier-transformed to R -space, and the curve fittings were performed in the R -space ($1.0\text{--}3.0 \text{ \AA}$). The amplitude reduction factor (S_0^2) for Dy–Cl bonds in this study was estimated to be 0.91. Here, the E_0 is 7786 eV.



Supplementary Figure 52. X-ray absorption spectra for DyCl₃ and DyCl₃@SWCNTs.



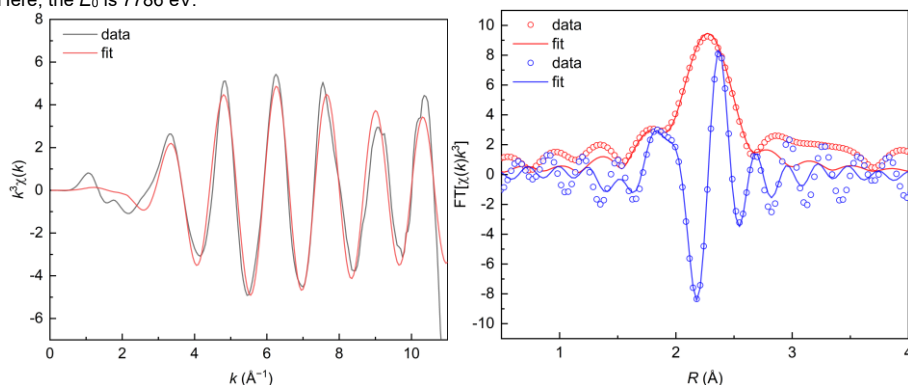
Supplementary Figure 53. Curve fitting results of k -weighted EXAFS oscillations (left) and Fourier transforms (right) at Dy L_3 -edge for DyCl₃. Black circles: observed; Red solid lines: fitted. Blue circles: imaginary parts; blue dotted lines: fitted.

Supplementary Table 15. Optimized parameters obtained from the curve fitting of k -weighted EXAFS oscillations for DyCl₃.

Shell	N	R (nm)	σ^2 (10^{-4} nm^2)	ΔE_0 (eV)	R -factor (%)
Dy-Cl	2	0.274 ± 0.003	4.7 ± 0.1	2.7 ± 0.9	0.6
Dy-Cl	4	0.263 ± 0.005			

5.2 Quenched DyCl₃

In the quenched DyCl₃ case, the extracted k^3 -weighted EXAFS oscillations over $k = 3\text{--}11 \text{ \AA}^{-1}$ after the smoothing background subtracted the XAFS spectra were Fourier-transformed to R -space, and the curve fittings were performed in the R -space ($1.0\text{--}3.0 \text{ \AA}$). The amplitude reduction factor (S_0^2) for Dy–Cl bonds in this study was estimated to be 0.91. Here, the E_0 is 7786 eV.



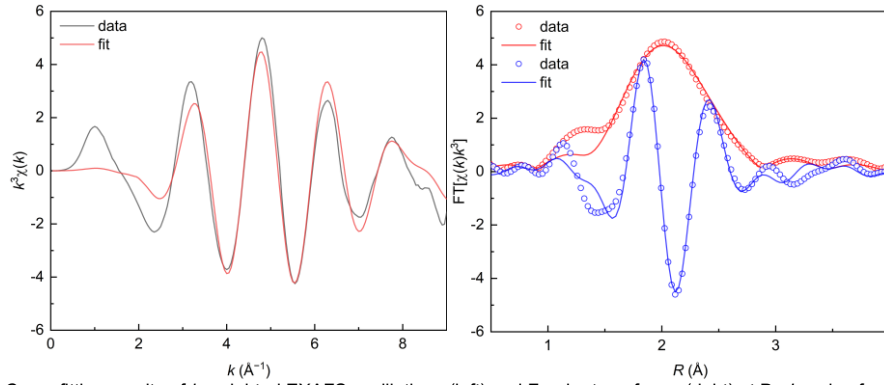
Supplementary Figure 54. Curve fitting results of k -weighted EXAFS oscillations (left) and Fourier transforms (right) at Dy L_3 -edge for DyCl₃ (quenched). Black circles: observed; Red solid lines: fitted. Blue circles: imaginary parts; blue dotted lines: fitted.

Supplementary Table 16. Optimized parameters obtained from the curve fitting of k -weighted EXAFS oscillations for DyCl₃ (quenched).

Shell	N	R (nm)	σ^2 (10^{-4} nm^2)	ΔE_0 (eV)	R -factor (%)
Dy-Cl	1.2 ± 0.5	0.251 ± 0.005	1.7 ± 3.0	-1.1 ± 2.9	4.3
Dy-Cl	3.7 ± 1.3	0.266 ± 0.002			

5.3 DyCl₃@SWCNTs

In the DyCl₃@SWCNTs case, the extracted k^3 -EXAFS oscillations over $k = 3\text{--}9\text{ \AA}^{-1}$ after the smoothing background subtracted the XAFS spectra were Fourier-transformed to R -space, and the curve fittings were performed in the R -space (1.0–3.0 Å). The amplitude reduction factor (S_0^2) for Dy–Cl bonds in this study was estimated to be 0.91. Here, the E_0 is 7786 eV.



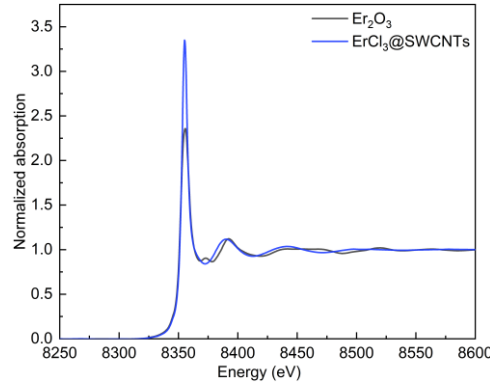
Supplementary Figure 55. Curve fitting results of k -weighted EXAFS oscillations (left) and Fourier transforms (right) at Dy L_3 -edge for DyCl₃@SWCNTs. Black circles: observed; Red solid lines: fitted. Blue circles: imaginary parts; blue dotted lines: fitted.

Supplementary Table 17. Optimized parameters obtained from the curve fitting of k -weighted EXAFS oscillations for DyCl₃@SWCNTs.

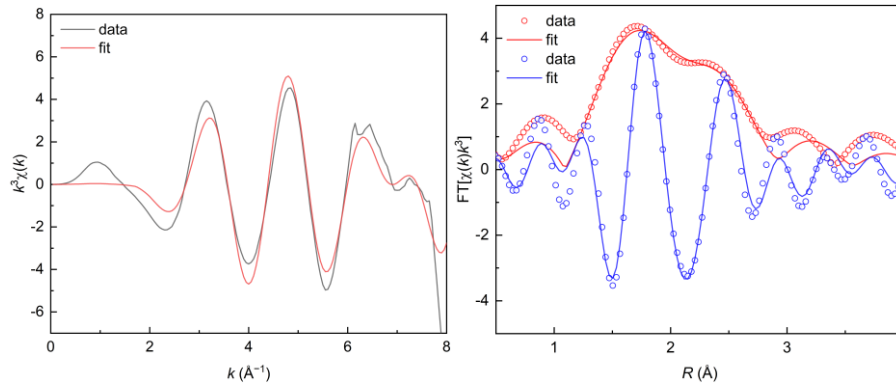
Shell	N	R (nm)	σ^2 (10^{-4} nm ²)	ΔE_0 (eV)	R -factor (%)
Dy-Cl	3.9 ± 0.7	0.271 ± 0.004	4.0 (fixed)	-3.7 ± 3.4	3.2
Dy-Cl	3.7 ± 0.6	0.252 ± 0.004			

5.4 ErCl₃@SWCNTs

In the ErCl₃@SWCNTs case, the extracted k^3 -weighted EXAFS oscillations over $k = 3\text{--}8\text{ \AA}^{-1}$ after the smoothing background subtracted the XAFS spectra were Fourier-transformed to R -space, and the curve fittings were performed in the R -space (1.0–3.0 Å). The amplitude reduction factor (S_0^2) for Er–Cl bonds in this study was estimated to be 0.94. Here, the E_0 is 8354 eV.



Supplementary Figure 56. X-ray absorption spectra for Er₂O₃ and ErCl₃@SWCNTs.



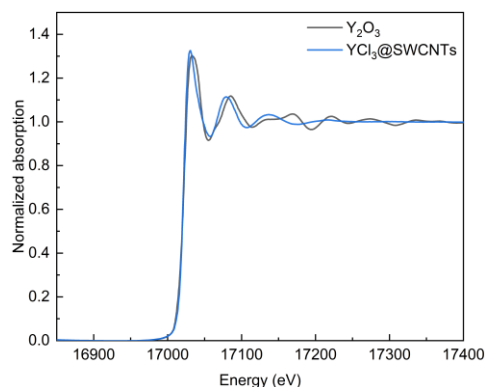
Supplementary Figure 57. Curve fitting results of k -weighted EXAFS oscillations (left) and Fourier transforms (right) at Er L_3 -edge for ErCl₃@SWCNTs. Black circles: observed; Red solid lines: fitted. Blue circles: imaginary parts; blue dotted lines: fitted.

Supplementary Table 18. Optimized parameters obtained from the curve fitting of k -weighted EXAFS oscillations for ErCl₃@SWCNTs.

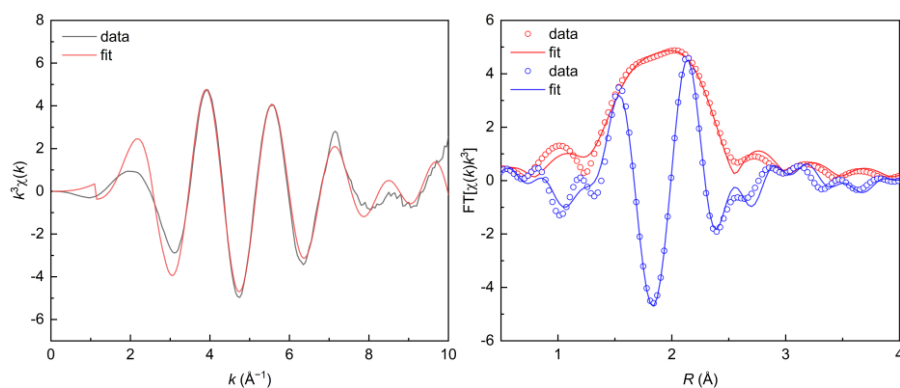
Shell	N	R (nm)	σ^2 (10^{-4} nm ²)	ΔE_0 (eV)	R -factor (%)
Er-Cl	4.0 ± 0.8	0.243 ± 0.006	0 (fixed)	-7.2 ± 5.6	4.0
Er-Cl	4.7 ± 1.0	0.265 ± 0.006			

5.5 YCl₃@SWCNTs

In the YCl₃@SWCNTs case, the extracted k^3 -weighted EXAFS oscillations over $k = 3\text{--}10 \text{ \AA}^{-1}$ after the smoothing background subtracted the XAFS spectra were Fourier-transformed to R -space, and the curve fittings were performed in the R -space ($1.0\text{--}3.0 \text{ \AA}$). The amplitude reduction factor (S_0^2) for Y–Cl bonds in this study was estimated to be 1.06. Here, the E_0 is 17022 eV.



Supplementary Figure 58. X-ray absorption spectra for Y₂O₃ and YCl₃@SWCNTs.

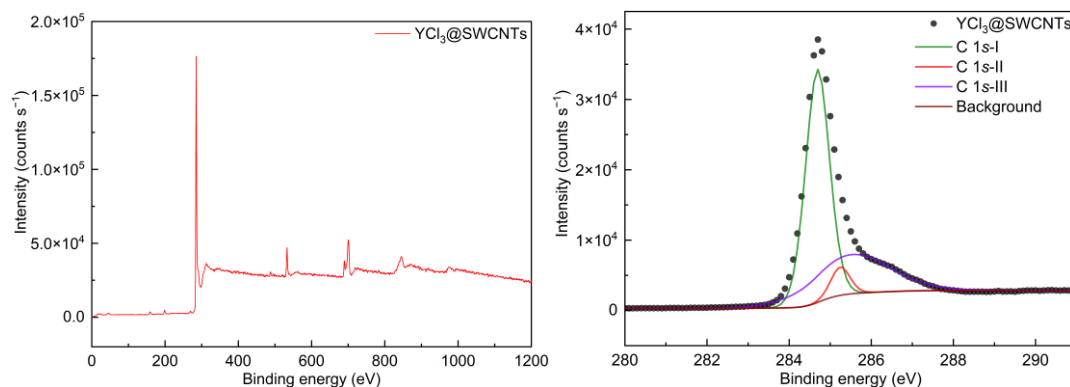


Supplementary Figure 59. Curve fitting results of k -weighted EXAFS oscillations (left) and Fourier transforms (right) at Y K -edge for YCl₃@SWCNTs. Black circles: observed; Red solid lines: fitted. Blue circles: imaginary parts; blue dotted lines: fitted.

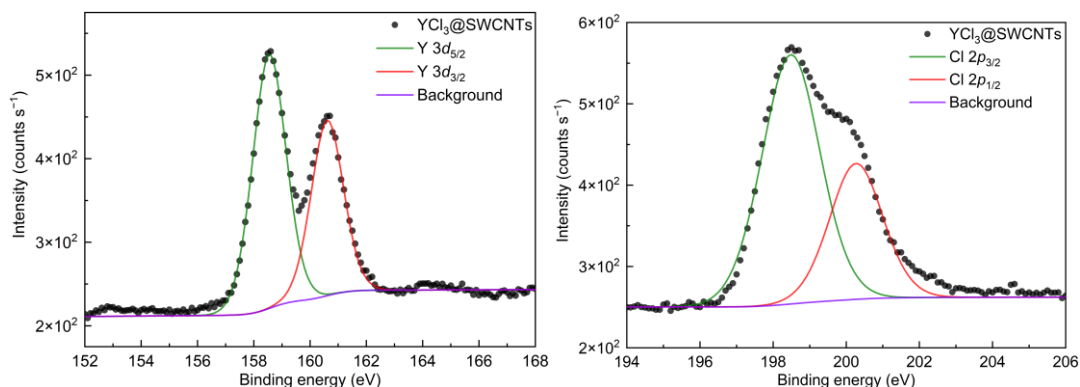
Supplementary Table 19. Optimized parameters obtained from the curve fitting of k -weighted EXAFS oscillations for YCl₃@SWCNTs.

Shell	N	R (nm)	σ^2 (10^{-4} nm^2)	ΔE_0 (eV)	R -factor (%)
Y-Cl	3.2 ± 0.3	0.234 ± 0.002	0 (fixed)	5.0 ± 1.7	1.8
Y-Cl	4.2 ± 0.4	0.252 ± 0.002			

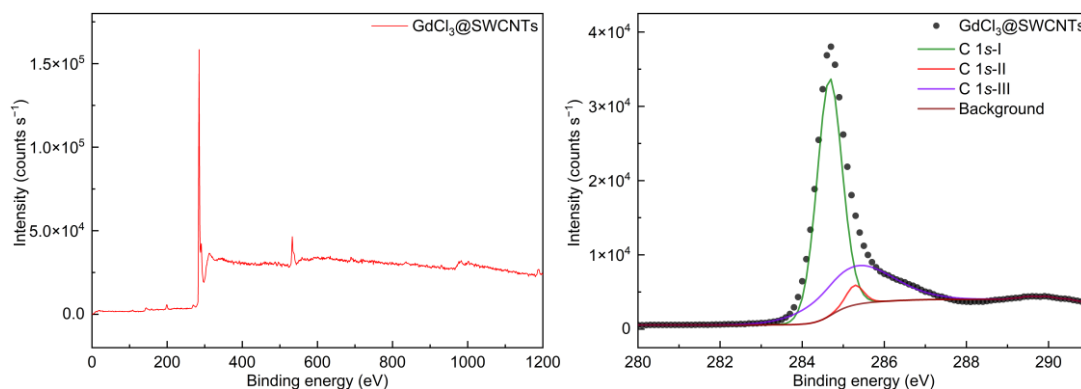
6 X-ray photoelectron spectroscopy (XPS)



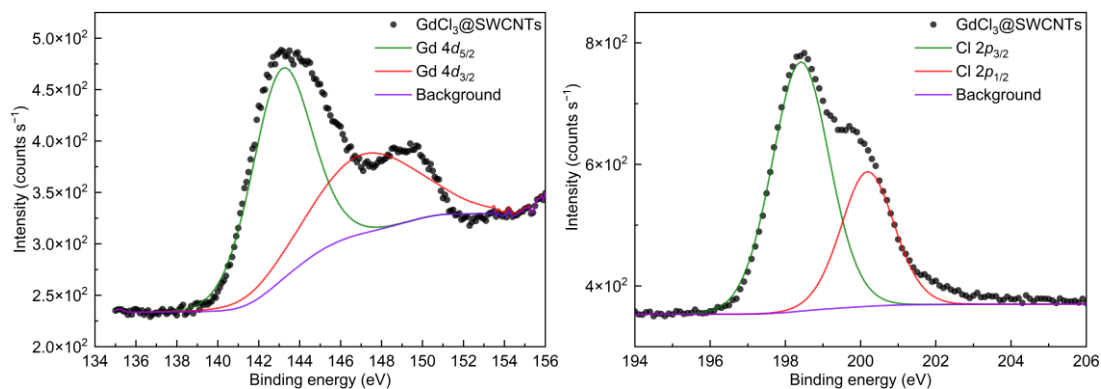
Supplementary Figure 60. The XPS wide region scan (left) and carbon 1s peak (right) of $\text{YCl}_3@\text{SWCNTs}$.



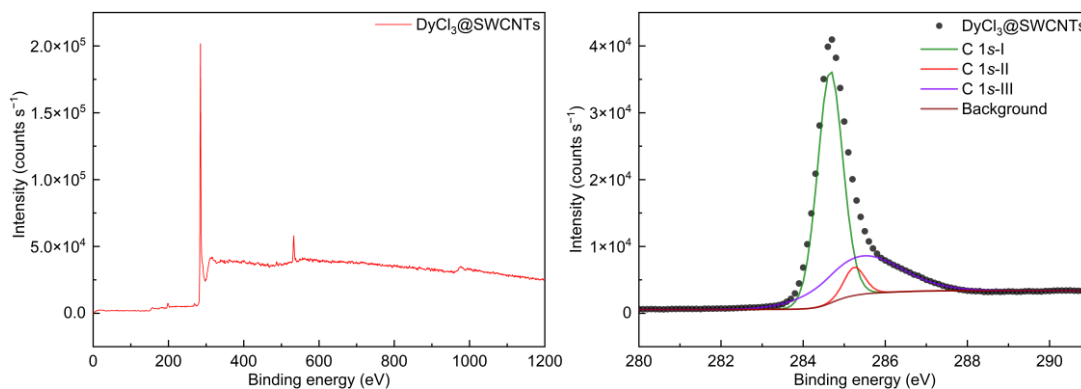
Supplementary Figure 61. Yttrium 3d peak (left) and chlorine 2p peak (right) on XPS spectrum of $\text{YCl}_3@\text{SWCNTs}$.



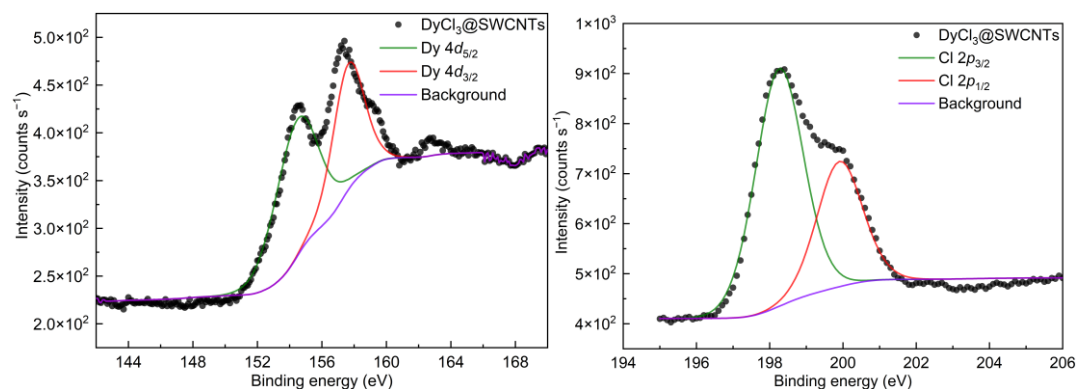
Supplementary Figure 62. The XPS wide region scan (left) and carbon 1s peak (right) of $\text{GdCl}_3@\text{SWCNTs}$.



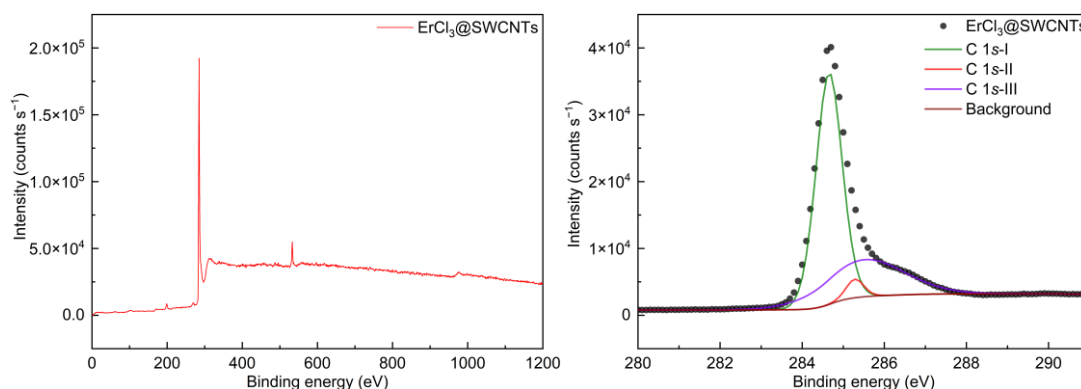
Supplementary Figure 63. Gadolinium 4d peak (left) and chlorine 2p peak (right) on XPS spectrum of $\text{GdCl}_3@\text{SWCNTs}$.



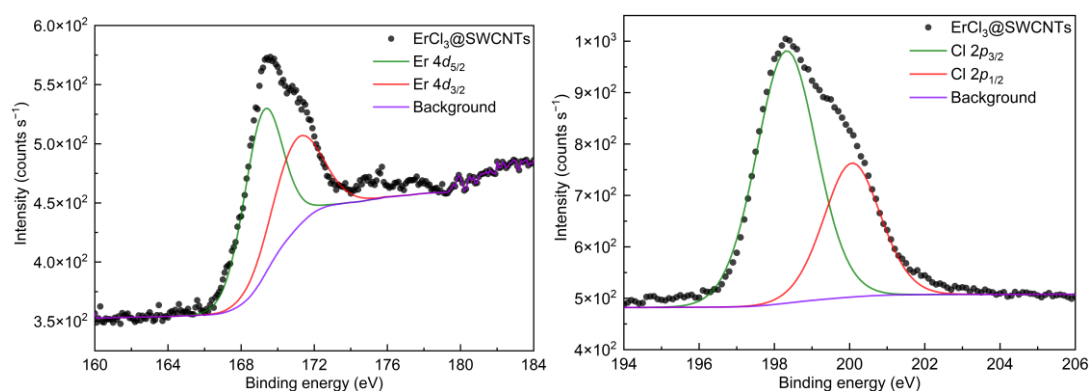
Supplementary Figure 64. The XPS wide region scan (left) and carbon 1s peak (right) of $\text{DyCl}_3@\text{SWCNTs}$.



Supplementary Figure 65. Dysprosium 4d peak (left) and chlorine 2p peak (right) on XPS spectrum of $\text{DyCl}_3@\text{SWCNTs}$.



Supplementary Figure 66. The XPS wide region scan (left) and carbon 1s peak (right) of $\text{ErCl}_3@\text{SWCNTs}$.



Supplementary Figure 67. Erbium 4d peak (left) and chlorine 2p peak (right) on XPS spectrum of $\text{ErCl}_3@\text{SWCNTs}$.

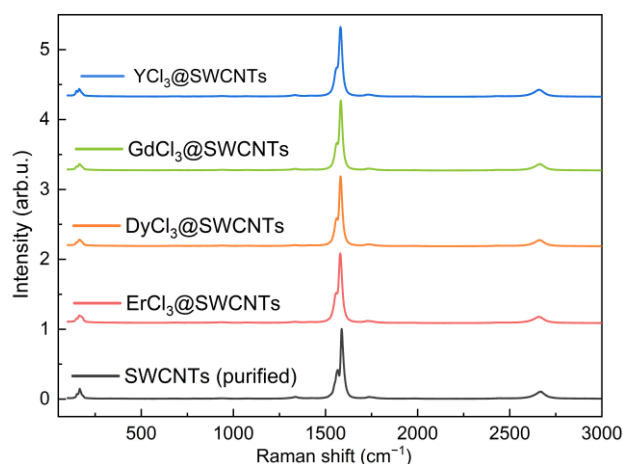
Supplementary Table 20. XPS peaks assignment and fitting result of purified SWCNTs, DyCl₃@SWCNTs, ErCl₃@SWCNTs, GdCl₃@SWCNTs, and YCl₃@SWCNTs.

SWCNTs (purified)							
Name				C 1s-I	C 1s-II	C 1s-III	
Position				284.74	284.89	285.52	
FWHM				0.48	0.73	2.32	
Area				18011.85	16819.20	13143.92	
YCl ₃ @SWCNTs							
Name	Y 3d _{5/2}	Y 3d _{3/2}	Cl 2p _{3/2}	Cl 2p _{1/2}	C 1s-I	C 1s-II	C 1s-III
Position	158.55	160.61	198.49	200.27	284.69	285.24	285.52
FWHM	1.40	1.35	1.87	1.71	0.68	0.54	2.23
Area	460.80	307.20	621.19	310.59	24538.64	2376.32	13501.64
GdCl ₃ @SWCNTs							
Name	Gd 4d _{5/2}	Gd 4d _{3/2}	Cl 2p _{3/2}	Cl 2p _{1/2}	C 1s-I	C 1s-II	C 1s-III
Position	143.10	147.13	198.41	200.19	284.67	285.28	285.31
FWHM	3.47	6.16	1.79	1.66	0.72	0.50	2.14
Area	772.09	514.73	797.70	398.85	24841.60	1419.72	12050.63
DyCl ₃ @SWCNTs							
Name	Dy 4d _{5/2}	Dy 4d _{3/2}	Cl 2p _{3/2}	Cl 2p _{1/2}	C 1s-I	C 1s-II	C 1s-III
Position	154.44	157.63	198.26	199.91	284.66	285.25	285.44
FWHM	3.18	2.30	1.54	1.46	0.72	0.58	2.26
Area	494.31	329.54	792.96	396.48	27164.52	2689.84	13999.84
ErCl ₃ @SWCNTs							
Name	Er 4d _{5/2}	Er 4d _{3/2}	Cl 2p _{3/2}	Cl 2p _{1/2}	C 1s-I	C 1s-II	C 1s-III
Position	169.18	171.03	198.32	200.06	284.65	285.28	285.52
FWHM	2.51	3.11	1.89	1.78	0.75	0.51	2.30
Area	382.86	255.24	1004.25	502.13	28110.21	1528.32	13794.48

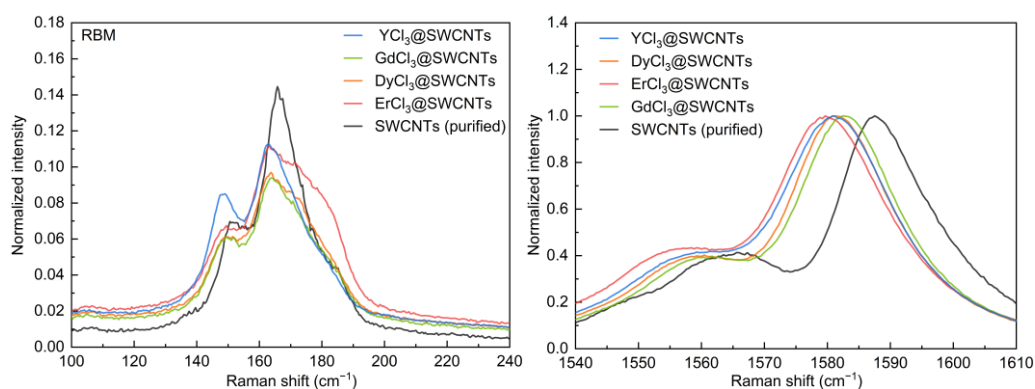
Supplementary Table 21. The atomic ratio of C/Ln/Cl calculated from XPS for DyCl₃@SWCNTs, ErCl₃@SWCNTs, GdCl₃@SWCNTs, and YCl₃@SWCNTs.

	$n(C)/n(Ln)$	$n(C)/n(Cl)$	$n(Cl)/n(Ln)$
YCl ₃ @SWCNTs	157.83	49.77	3.17
GdCl ₃ @SWCNTs	155.38	36.74	4.23
DyCl ₃ @SWCNTs	305.38	42.30	7.22
ErCl ₃ @SWCNTs	429.06	33.08	12.97

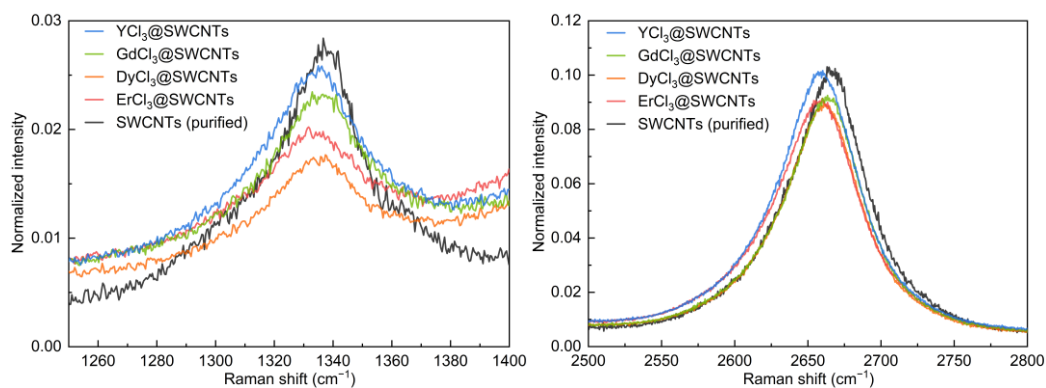
7 Raman spectroscopy



Supplementary Figure 68. The wide region Raman spectrum for purified SWCNTs, DyCl₃@SWCNTs, ErCl₃@SWCNTs, GdCl₃@SWCNTs, and YCl₃@SWCNTs respectively. The intensities were normalized to their respective G-band intensities.



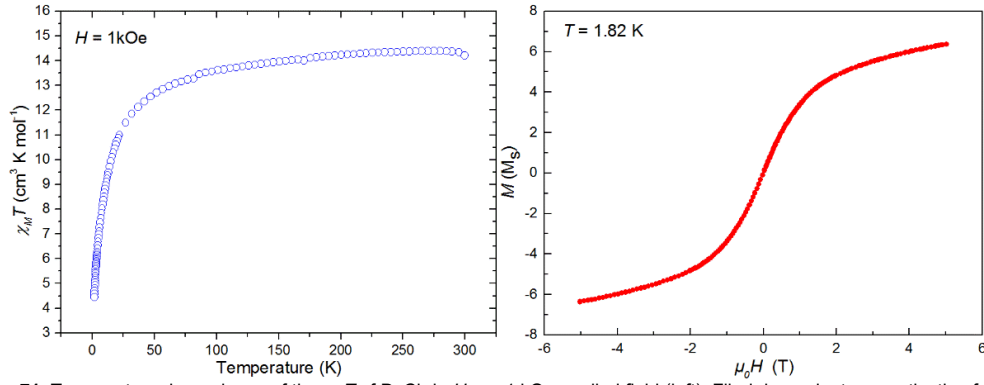
Supplementary Figure 69. The radial breathing mode (RBM) (left) and G band (right) region Raman spectrum for purified SWCNT, DyCl₃@SWCNTs, ErCl₃@SWCNTs, GdCl₃@SWCNTs, and YCl₃@SWCNTs respectively. The intensities were normalized to their respective G-band intensities.



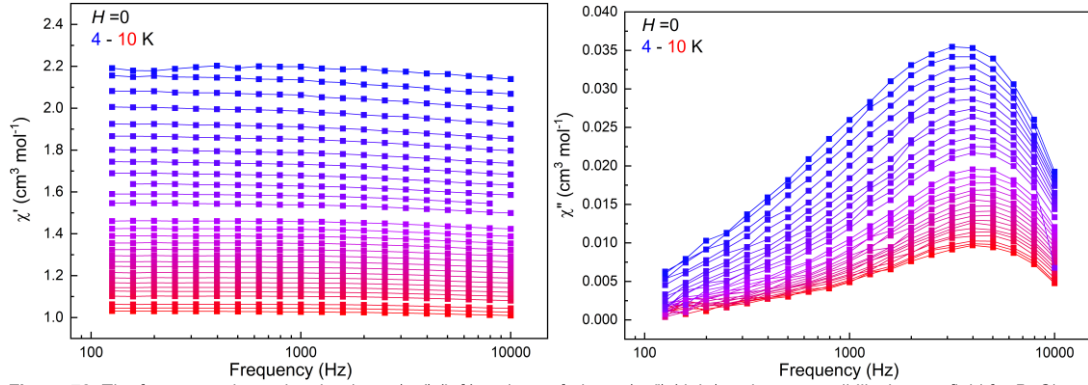
Supplementary Figure 70. The D band (left) and 2D band (right) region Raman spectrum for purified SWCNT, DyCl₃@SWCNTs, ErCl₃@SWCNTs, GdCl₃@SWCNTs, and YCl₃@SWCNTs respectively. The intensities were normalized to their respective G-band intensities.

8 Magnetic properties

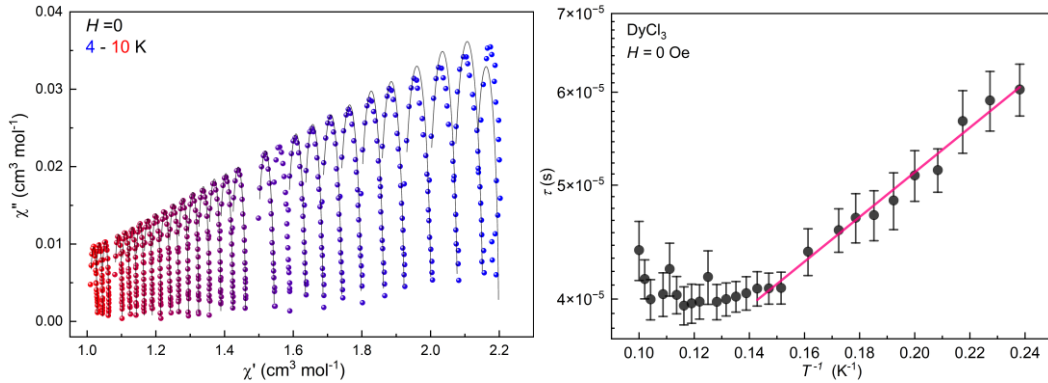
8.1 Crystalline DyCl₃ & Quenched DyCl₃



Supplementary Figure 71. Temperature dependence of the $\chi_m T$ of DyCl₃ in $H_{DC} = 1$ kOe applied field (left). Filed dependent magnetization for DyCl₃ ant 1.82K (right).



Supplementary Figure 72. The frequency dependent in-phase (χ'_m) (left) and out-of-phase (χ''_m) (right) molar susceptibility in zero field for DyCl₃.



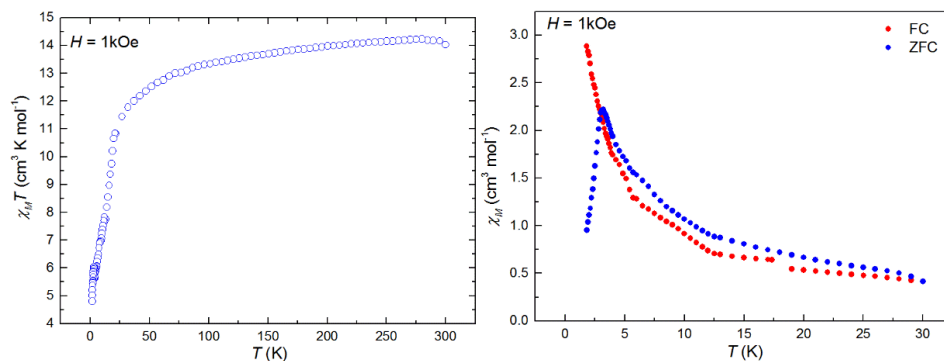
Supplementary Figure 73. Cole-Cole plot for DyCl₃ in a zero field (left). Relaxation times of the magnetization $\ln(\tau)$ vs T^{-1} in a zero field of DyCl₃ (black circle). Error bars indicate the standard uncertainty of the fitted parameter τ obtained by fitting 20 complex-valued data points to Eqs. (6), (7) with 4 real-valued free parameters. The magenta curve represents the fitting result obtained using Eq. (3).

Supplementary Table 22. The fitting parameter from ac susceptibility of DyCl₃ in a zero field by the Debye model.

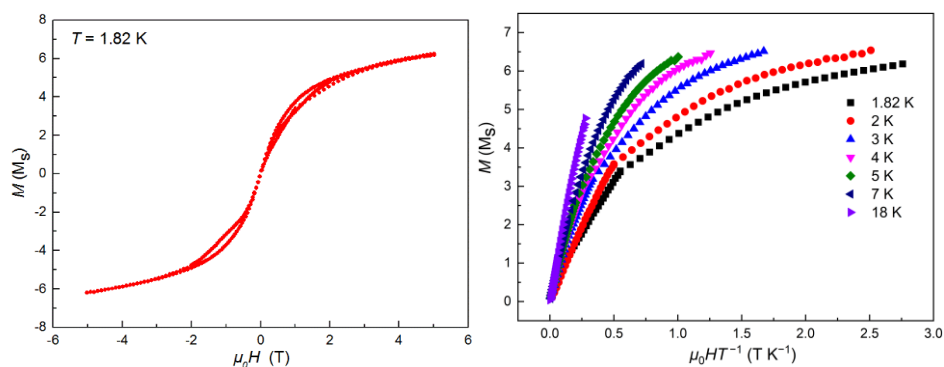
DyCl ₃ ($H = 0$)						
Temperature (K)	τ (s)	τ (error)	α	α (error)	Reduced Chi-Sqr	Adj. R-Square
4	3.91E-05	6.42E-06	6.76E-02	8.84E-02	5.25E-05	1.00E+00
4.2	6.03E-05	3.07E-06	1.52E-01	2.69E-02	7.00E-06	1.00E+00
4.4	5.90E-05	3.45E-06	1.53E-01	3.07E-02	8.40E-06	1.00E+00
4.6	5.67E-05	3.47E-06	1.58E-01	3.14E-02	7.72E-06	1.00E+00
4.8	5.15E-05	2.18E-06	1.34E-01	2.21E-02	3.28E-06	1.00E+00
5	5.10E-05	2.53E-06	1.32E-01	2.59E-02	4.13E-06	1.00E+00
5.2	4.85E-05	2.70E-06	1.38E-01	2.81E-02	4.20E-06	1.00E+00
5.4	4.72E-05	2.31E-06	1.25E-01	2.52E-02	3.04E-06	1.00E+00
5.6	4.69E-05	2.26E-06	1.17E-01	2.51E-02	2.75E-06	1.00E+00
5.8	4.58E-05	1.95E-06	1.00E-01	2.33E-02	2.03E-06	1.00E+00
6	2.76E-05	2.65E-05	0.00E+00	5.09E-01	1.39E-02	9.77E-01
6.2	4.39E-05	2.01E-06	9.65E-02	2.43E-02	1.88E-06	1.00E+00
6.6	4.09E-05	1.28E-06	4.64E-02	1.79E-02	8.17E-07	1.00E+00
6.8	4.08E-05	1.24E-06	4.73E-02	1.74E-02	7.07E-07	1.00E+00

Supplementary Table 23. The fitting parameter from temperature scan ac susceptibility for DyCl₃ in a zero field.

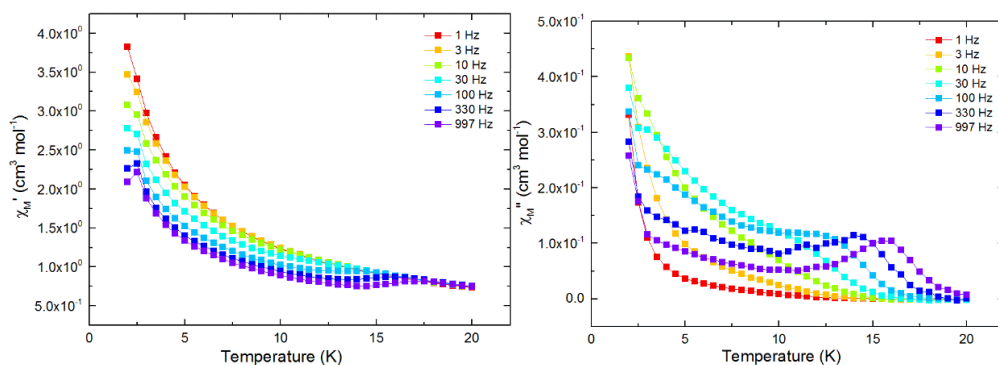
DyCl ₃ ($H = 0$)	
τ_0 (s)	$2.13531\text{E-}5 \pm 7.61101\text{E-}7$
U_{eff} (K)	4.38079 ± 0.18106
U_{eff} (cm ⁻¹)	3.04478 ± 0.12584
τ_{QTM} (s)	null
C	null
n	null
Reduced Chi-Sqr	9.18365E-13
R-Square (COD)	0.98132
Adj. R-Square	0.97963



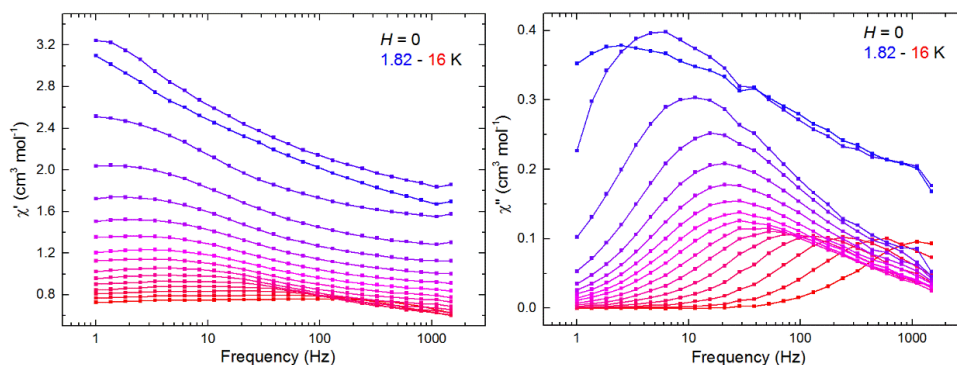
Supplementary Figure 74. Temperature dependence of the $\chi_m T$ of DyCl₃ (quenched) in a $H_{DC} = 1$ kOe applied field (left). Zero-field cooling (blue) and field cooling (red) curves of DyCl₃ (quenched) under a 1 kOe applied field.



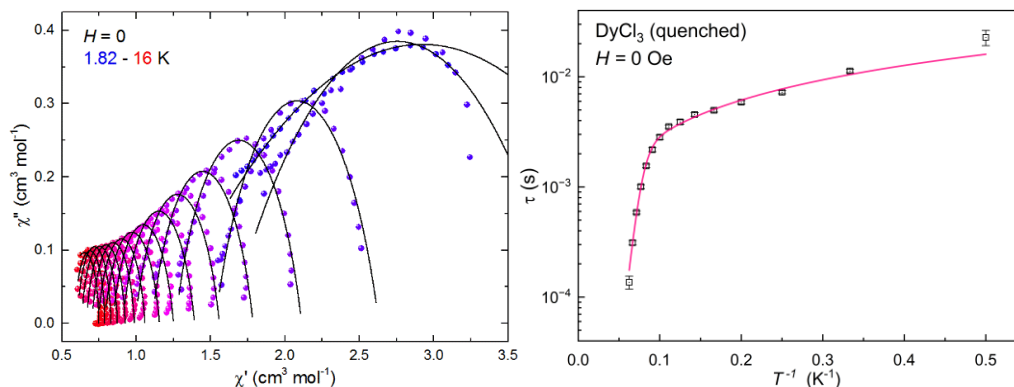
Supplementary Figure 75. Filed dependent magnetization for DyCl₃ (quenched) at 1.82K (left). Temperature reduced filed dependent magnetization for DyCl₃ (quenched) (right).



Supplementary Figure 76. The temperature dependent in-phase (χ_M') (left) and out-of-phase (χ_M'') (right) molar susceptibility in a zero field for DyCl₃ (quenched).



Supplementary Figure 77. The frequency dependent in-phase (χ_M') (left) and out-of-phase (χ_M'') (right) molar susceptibility in a zero field for DyCl₃ (quenched).



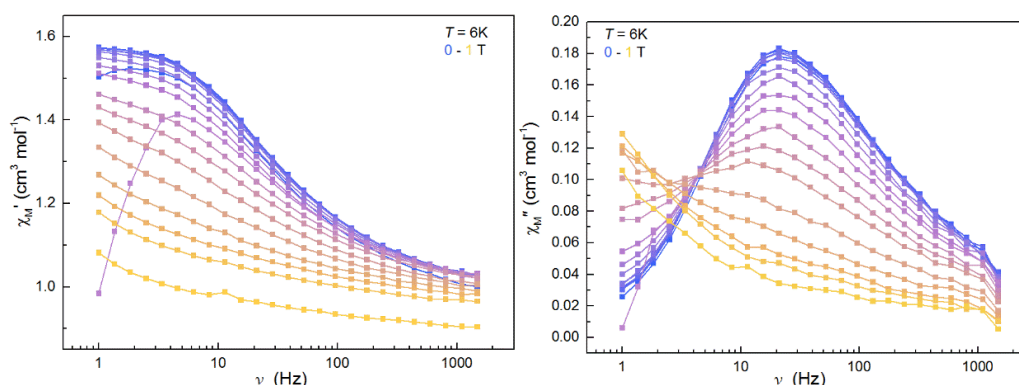
Supplementary Figure 78. Cole-Cole plot for DyCl₃ (quenched) in a zero field (left). Relaxation times of the magnetization $\ln(\tau)$ vs T^{-1} in a zero field of DyCl₃ (quenched) (black circle). Error bars indicate the standard uncertainty of the fitted parameter τ obtained by fitting 25 complex-valued data points to Eqs. (6), (7) with 4 real-valued free parameters. The magenta curve represents the fitting result obtained using Eq. (8).

Supplementary Table 24. The fitting parameter from ac susceptibility of DyCl₃ (quenched) in a zero field by the Debye model.

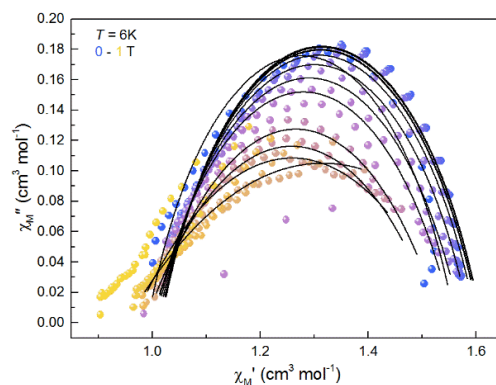
DyCl ₃ (quenched) ($H = 0$)						
Temperature (K)	τ (s)	τ (error)	α	α (error)	Reduced Chi-Sqr	Adj. R-Square
1.82	9.03E-02	2.08E-02	7.23E-01	1.16E-02	1.27E-04	1.00E+00
2	2.28E-02	3.60E-03	5.84E-01	2.30E-02	9.32E-04	9.99E-01
3	1.13E-02	4.19E-04	3.59E-01	1.28E-02	2.20E-04	1.00E+00
4	7.24E-03	2.41E-04	3.23E-01	1.29E-02	1.58E-04	1.00E+00
5	5.87E-03	2.05E-04	3.02E-01	1.42E-02	1.31E-04	1.00E+00
6	4.96E-03	1.89E-04	2.91E-01	1.59E-02	1.18E-04	1.00E+00
7	4.54E-03	1.66E-04	2.83E-01	1.54E-02	8.30E-05	1.00E+00
8	3.89E-03	1.77E-04	2.59E-01	2.00E-02	1.06E-04	1.00E+00
9	3.53E-03	1.32E-04	2.46E-01	1.69E-02	6.32E-05	1.00E+00
10	2.83E-03	1.26E-04	1.94E-01	2.17E-02	8.47E-05	1.00E+00
11	2.18E-03	9.23E-05	1.64E-01	2.16E-02	7.18E-05	1.00E+00
12	1.55E-03	5.72E-05	1.21E-01	1.99E-02	5.23E-05	1.00E+00
13	1.01E-03	4.12E-05	7.91E-02	2.33E-02	5.99E-05	1.00E+00
14	5.87E-04	2.09E-05	3.80E-02	2.08E-02	3.78E-05	1.00E+00
15	3.12E-04	1.48E-05	0.00E+00	2.59E-02	3.54E-05	1.00E+00
16	1.36E-04	1.85E-05	0.00E+00	4.55E-02	4.29E-05	1.00E+00

Supplementary Table 25. The fitting parameter from temperature scan ac susceptibility for DyCl₃ (quenched) in a zero field.

DyCl ₃ (quenched) ($H = 0$)	
τ_0 (s)	$1.3464\text{E-}8 \pm 1.06751\text{E-}8$
U_{eff} (K)	153.24337 ± 11.15535
U_{eff} (cm ⁻¹)	106.50878 ± 7.75331
τ_{QTM} (s)	null
C	30.21311 ± 3.12876
n	1.0471 ± 0.05845
Reduced Chi-Sqr	2.60343
R-Square (COD)	0.99518
Adj. R-Square	0.99387



Supplementary Figure 79. The frequency dependent in-phase (χ'_M) (left) and out-of-phase (χ''_M) (right) molar susceptibility at 6 K under various applied field for DyCl₃ (quenched).

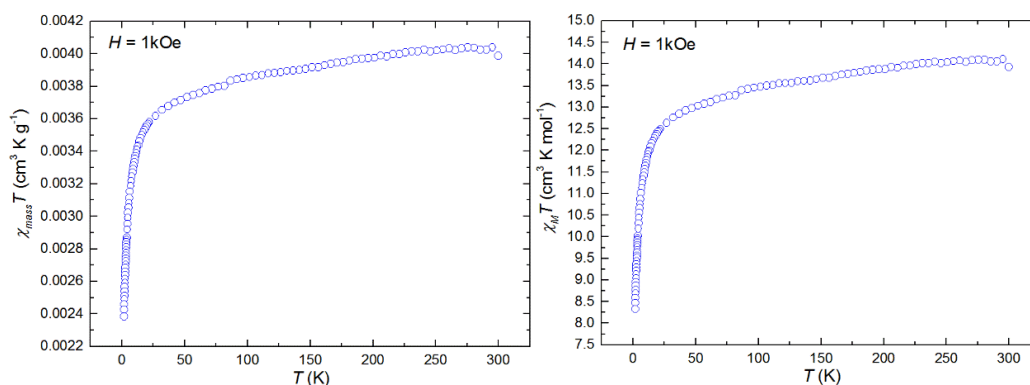


Supplementary Figure 80. The Cole-Cole plot for DyCl₃ (quenched) at 6 K under various applied field.

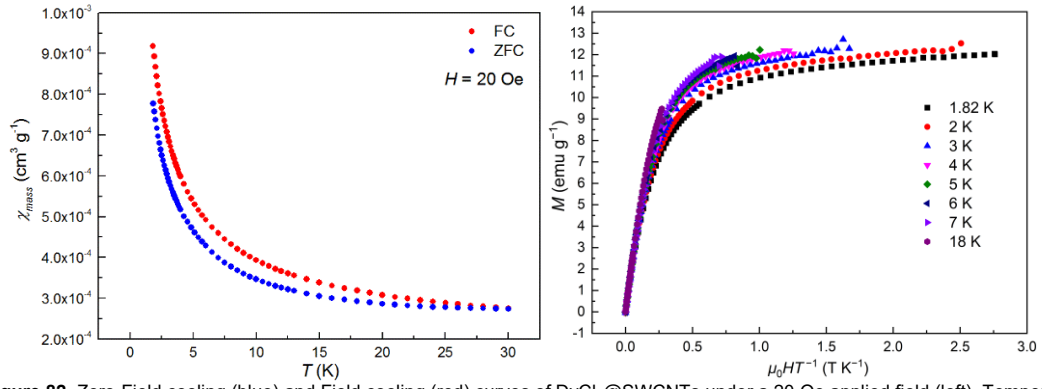
Supplementary Table 26. The fitting parameter from ac susceptibility of DyCl₃ (quenched) at 6 K under various field by the Debye model.

DyCl ₃ (quenched) (T = 6 K)						
Field (Oe)	τ (s)	τ (error)	α	α (error)	Reduced Chi-Sqr	Adj. R-Square
0	4.92E-03	1.84E-04	2.96E-01	1.52E-02	1.16E-04	1.00E+00
50	5.30E-03	1.59E-04	3.01E-01	1.21E-02	7.89E-05	1.00E+00
100	5.37E-03	1.64E-04	3.02E-01	1.23E-02	8.02E-05	1.00E+00
200	5.35E-03	1.61E-04	3.05E-01	1.21E-02	7.65E-05	1.00E+00
300	5.46E-03	1.55E-04	3.06E-01	1.14E-02	6.75E-05	1.00E+00
400	5.46E-03	1.57E-04	3.13E-01	1.14E-02	6.54E-05	1.00E+00
600	5.67E-03	1.52E-04	3.25E-01	1.03E-02	5.02E-05	1.00E+00
800	5.91E-03	1.50E-04	3.43E-01	9.38E-03	3.81E-05	1.00E+00
1000	6.36E-03	1.36E-04	3.63E-01	7.57E-03	2.23E-05	1.00E+00
1250	2.87E-03	8.92E-04	1.33E-01	1.66E-01	5.19E-03	9.84E-01
1500	8.63E-03	2.14E-04	4.37E-01	7.19E-03	1.41E-05	1.00E+00
1750	1.16E-02	3.47E-04	4.83E-01	7.16E-03	1.11E-05	1.00E+00
2000	2.13E-02	1.26E-03	5.51E-01	9.42E-03	1.45E-05	1.00E+00
2500	1.18E-01	2.44E-02	6.42E-01	1.28E-02	1.45E-05	1.00E+00
3000	1.49E+01	1.31E+01	7.28E-01	1.16E-02	5.71E-06	1.00E+00
3500	5.63E+06	1.29E+08	7.42E-01	1.58E-02	7.55E-06	1.00E+00
4000	1.49E+07	1.04E+09	6.96E-01	2.17E-02	1.48E-05	1.00E+00
6000	5.53E+07	9.17E+09	6.91E-01	3.23E-02	2.14E-05	1.00E+00

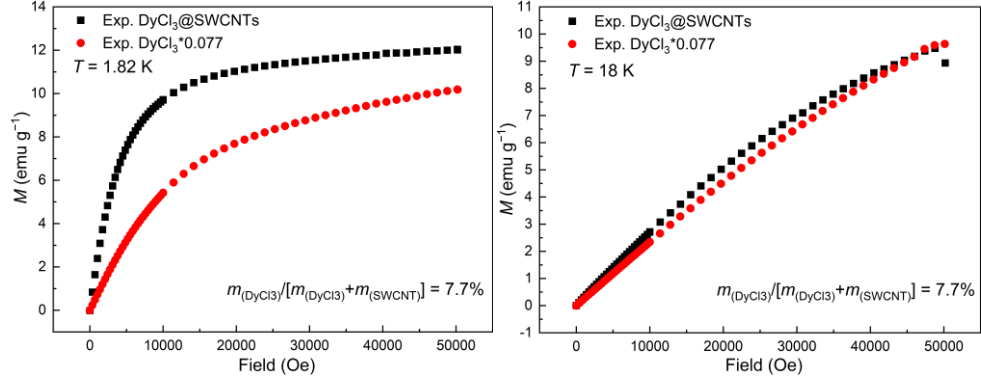
8.2 DyCl₃@SWCNTs



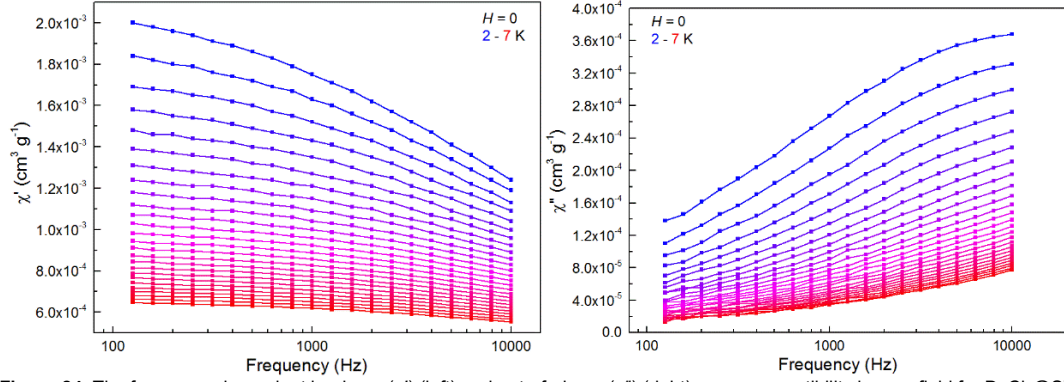
Supplementary Figure 81. Temperature dependence of the $\chi_{\text{mass}}T$ (left) and $\chi_{\text{mole}}T$ (right, normalized by Dy atom number which determined from M vs H data) of DyCl₃@SWCNTs in a $H_{\text{DC}} = 1$ kOe applied field.



Supplementary Figure 82. Zero-Field cooling (blue) and Field cooling (red) curves of DyCl₃@SWCNTs under a 20 Oe applied field (left). Temperature reduced field dependent magnetization for DyCl₃@SWCNTs (right).



Supplementary Figure 83. The mass magnetization comparison between DyCl₃@SWCNTs (black) and DyCl₃ (red) at 1.82 K (left) and 18 K (right).



Supplementary Figure 84. The frequency dependent in-phase (χ') (left) and out-of-phase (χ'') (right) mass susceptibility in zero field for DyCl₃@SWCNTs.

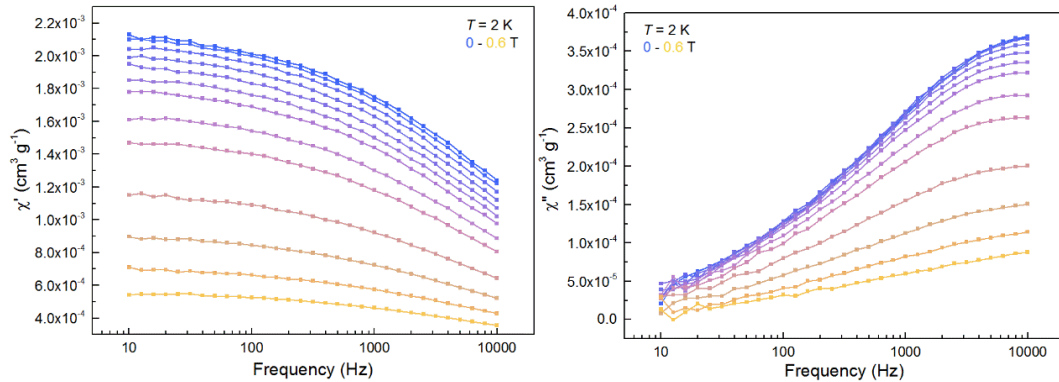
Supplementary Table 27. The fitting parameter from ac susceptibility of DyCl₃@SWCNTs in a zero field by the Debye model.

DyCl ₃ @SWCNTs ($H = 0$)						
Temperature (K)	τ (s)	τ (error)	α	α (error)	Reduced Chi-Sqr	Adj. R-Square
2	1.38E-05	5.69E-07	5.42E-01	4.22E-03	6.14E-12	1.00E+00
2.2	1.14E-05	4.29E-07	5.37E-01	3.59E-03	3.35E-12	1.00E+00
2.4	9.41E-06	3.96E-07	5.35E-01	3.71E-03	2.74E-12	1.00E+00
2.6	7.91E-06	3.89E-07	5.35E-01	4.03E-03	2.51E-12	1.00E+00
2.8	6.78E-06	3.87E-07	5.36E-01	4.38E-03	2.33E-12	1.00E+00
3	5.36E-06	3.65E-07	5.42E-01	4.67E-03	2.08E-12	1.00E+00
3.2	4.54E-06	3.58E-07	5.44E-01	5.04E-03	1.96E-12	1.00E+00
3.4	3.91E-06	3.67E-07	5.44E-01	5.66E-03	2.03E-12	1.00E+00
3.6	2.31E-06	4.46E-07	5.70E-01	9.09E-03	3.92E-12	1.00E+00
3.8	2.33E-06	3.89E-07	5.61E-01	8.04E-03	2.71E-12	1.00E+00
4	1.90E-06	3.03E-07	5.67E-01	7.02E-03	1.71E-12	1.00E+00
4.2	1.73E-06	2.91E-07	5.67E-01	7.18E-03	1.54E-12	1.00E+00
4.4	1.57E-06	3.08E-07	5.66E-01	8.08E-03	1.68E-12	1.00E+00
4.6	1.43E-06	4.07E-07	5.66E-01	1.14E-02	2.88E-12	1.00E+00
4.8	1.22E-06	6.23E-07	5.72E-01	1.91E-02	6.73E-12	1.00E+00
5	1.15E-06	5.79E-07	5.67E-01	1.86E-02	5.68E-12	1.00E+00
5.2	1.06E-06	6.21E-07	5.66E-01	2.10E-02	6.38E-12	1.00E+00
5.4	9.75E-07	6.81E-07	5.66E-01	2.43E-02	7.56E-12	1.00E+00

5.6	9.99E-07	4.47E-07	5.64E-01	1.58E-02	3.03E-12	1.00E+00
5.8	9.49E-07	4.27E-07	5.62E-01	1.56E-02	2.69E-12	1.00E+00
6	9.17E-07	4.56E-07	5.54E-01	1.72E-02	3.00E-12	1.00E+00
6.2	8.74E-07	5.21E-07	5.53E-01	2.03E-02	3.79E-12	1.00E+00
6.4	8.18E-07	4.85E-07	5.56E-01	1.97E-02	3.21E-12	1.00E+00
6.6	7.91E-07	4.85E-07	5.51E-01	2.02E-02	3.12E-12	1.00E+00
6.8	7.43E-07	5.16E-07	5.53E-01	2.23E-02	3.46E-12	1.00E+00
7	7.28E-07	5.33E-07	5.49E-01	2.34E-02	3.56E-12	1.00E+00

Supplementary Table 28. The fitting parameter from temperature scan ac susceptibility for DyCl₃@SWCNTs in zero field.

DyCl ₃ @SWCNTs ($H = 0$)	
τ_0 (s)	$8.28751\text{E-}8 \pm 1.23428\text{E-}8$
U_{eff} (K)	13.59649 ± 0.51898
U_{eff} (cm ⁻¹)	9.44997 ± 0.36071
τ_{QTM} (s)	$1.63638\text{E-}5 \pm 7.23508\text{E-}7$
C	0 ± 0
n	9 ± 0
Reduced Chi-Sqr	0.33513
R-Square (COD)	0.9951
Adj. R-Square	0.99467



Supplementary Figure 85. The frequency dependent in-phase (χ') (left) and out-of-phase (χ'') (right) mass susceptibility at 2 K under various applied field for DyCl₃@SWCNT.

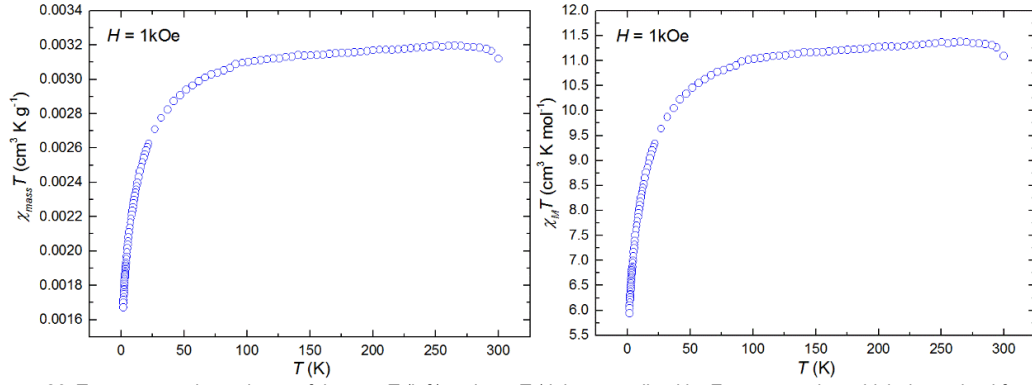
Supplementary Table 29. The fitting parameter from ac susceptibility of for DyCl₃@SWCNTs at 2 K under various field by the Debye model.

DyCl ₃ @SWCNTs ($T = 2$ K)						
Field (Oe)	τ (s)	τ (error)	α	α (error)	Reduced Chi-Sqr	Adj. R-Square
0	1.57E-05	8.26E-07	5.28E-01	4.52E-03	2.03E-11	1.00E+00
199	1.59E-05	7.94E-07	5.29E-01	4.27E-03	1.85E-11	1.00E+00
399	1.74E-05	8.23E-07	5.24E-01	4.27E-03	1.88E-11	1.00E+00
600	1.81E-05	7.16E-07	5.23E-01	3.63E-03	1.33E-11	1.00E+00
799	1.82E-05	6.83E-07	5.29E-01	3.38E-03	1.10E-11	1.00E+00
999	2.02E-05	1.00E-06	5.18E-01	4.82E-03	2.16E-11	1.00E+00
1200	1.99E-05	8.92E-07	5.26E-01	4.22E-03	1.53E-11	1.00E+00
1600	2.09E-05	1.11E-06	5.24E-01	5.10E-03	1.89E-11	1.00E+00
1999	2.20E-05	1.13E-06	5.21E-01	5.11E-03	1.57E-11	1.00E+00
3000	1.78E-05	9.94E-07	5.53E-01	4.61E-03	6.82E-12	1.00E+00
4000	1.60E-05	1.56E-06	5.58E-01	7.62E-03	9.83E-12	1.00E+00
5000	1.08E-05	2.21E-06	5.78E-01	1.31E-02	1.44E-11	1.00E+00
6000	8.54E-06	2.14E-06	5.78E-01	1.48E-02	9.89E-12	1.00E+00

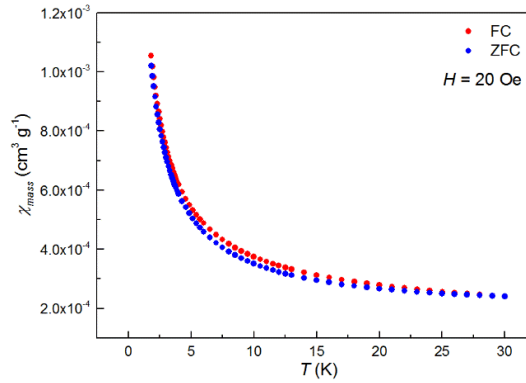
Supplementary Table 30. The fitting parameter of the relaxation process from field scan ac susceptibility for DyCl₃@SWCNTs at 2 K.

DyCl ₃ @SWCNTs ($T = 2$ K)	
a	$6.661\text{E-}11 \pm 7.8133\text{E-}12$
B	$64020.13255 \pm 2668.30344$
C	$1.91064\text{E-}6 \pm 1.11313\text{E-}6$
D	$2.67661\text{E-}6 \pm 1.53779\text{E-}6$
Reduced Chi-Sqr	7.21E-13
R-Square (COD)	0.96318
Adj. R-Square	0.9509

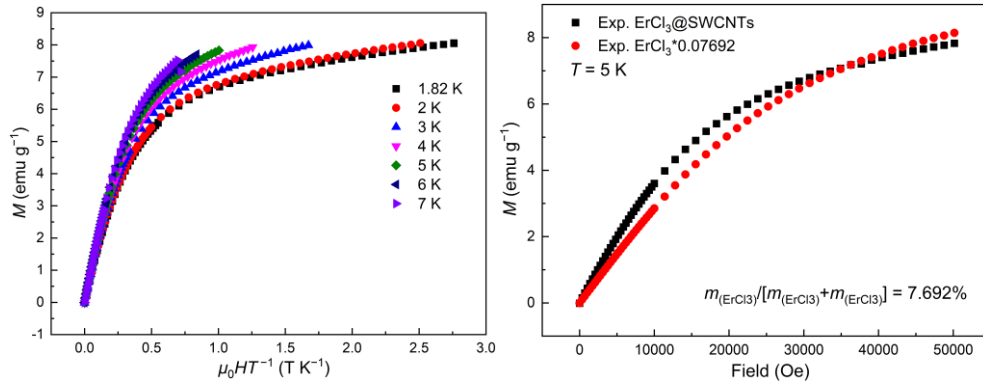
8.3 ErCl₃@SWCNTs & GdCl₃@SWCNTs



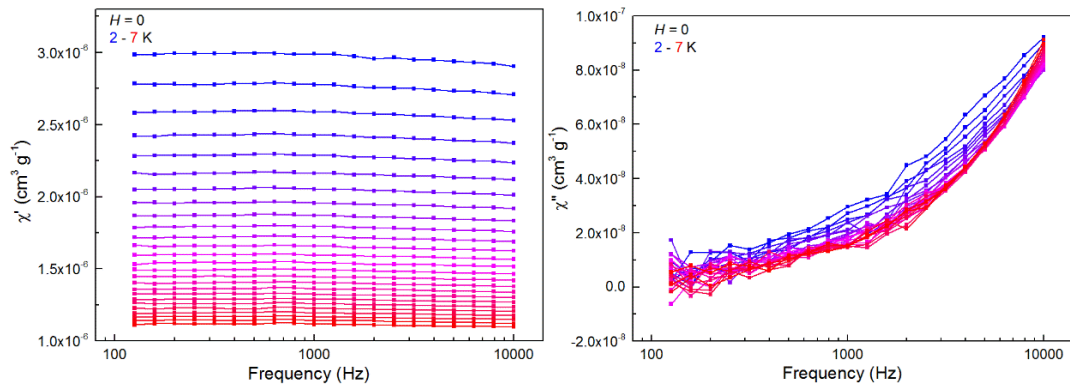
Supplementary Figure 86. Temperature dependence of the $\chi_{\text{mass}} T$ (left) and $\chi_{\text{mole}} T$ (right, normalized by Er atom number which determined from M vs H data) of ErCl₃@SWCNTs in a $H_{\text{DC}} = 1 \text{ kOe}$ applied field.



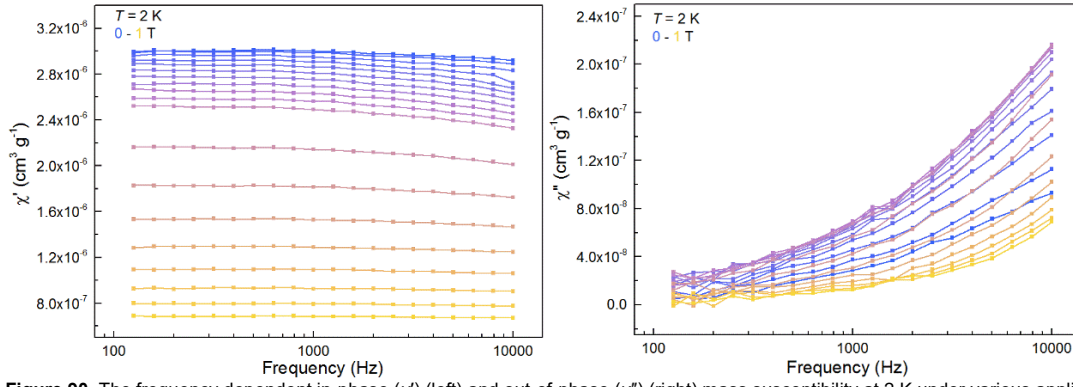
Supplementary Figure 87. Zero-Field cooling (blue) and Field cooling (red) curves of ErCl₃@SWCNTs under a 20 Oe applied field.



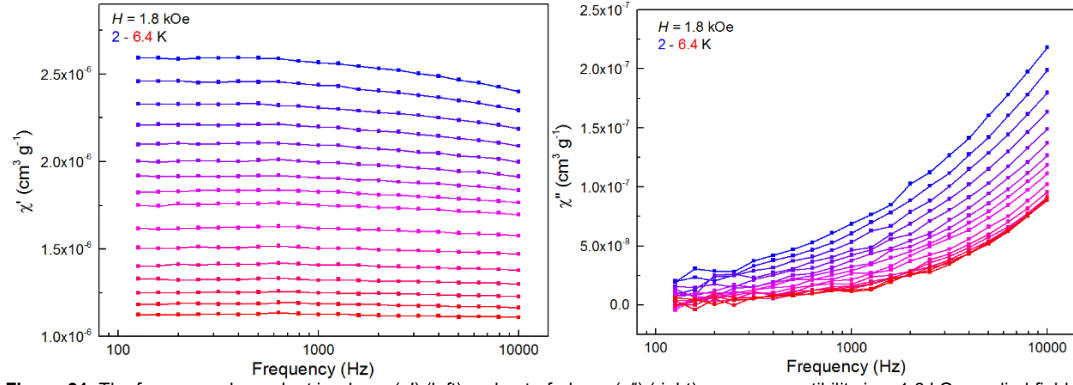
Supplementary Figure 88. Temperature reduced field dependent magnetization for ErCl₃@SWCNTs (left). The mass magnetization comparison between ErCl₃@SWCNTs (black) and ErCl₃ (red) at 5 K (right).



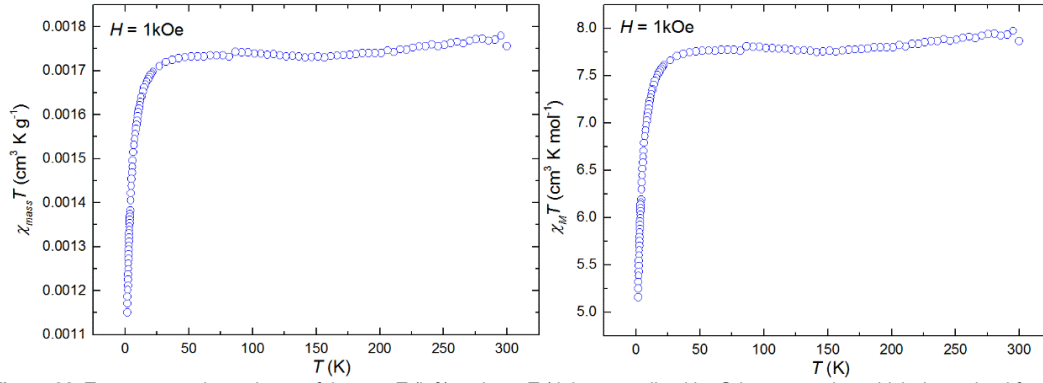
Supplementary Figure 89. The frequency dependent in-phase (χ') (left) and out-of-phase (χ'') (right) mass susceptibility in zero field for ErCl₃@SWCNTs.



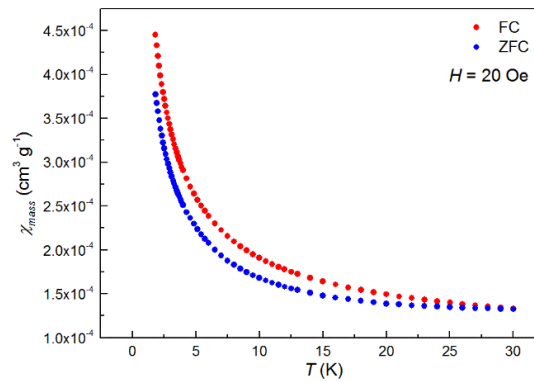
Supplementary Figure 90. The frequency dependent in-phase (χ') (left) and out-of-phase (χ'') (right) mass susceptibility at 2 K under various applied field for $\text{ErCl}_3\text{@SWCNTs}$.



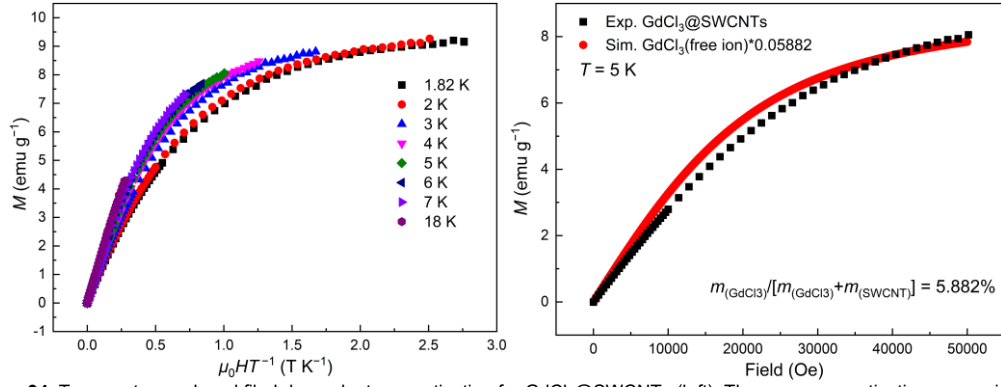
Supplementary Figure 91. The frequency dependent in-phase (χ') (left) and out-of-phase (χ'') (right) mass susceptibility in a 1.8 kOe applied field for $\text{ErCl}_3\text{@SWCNTs}$.



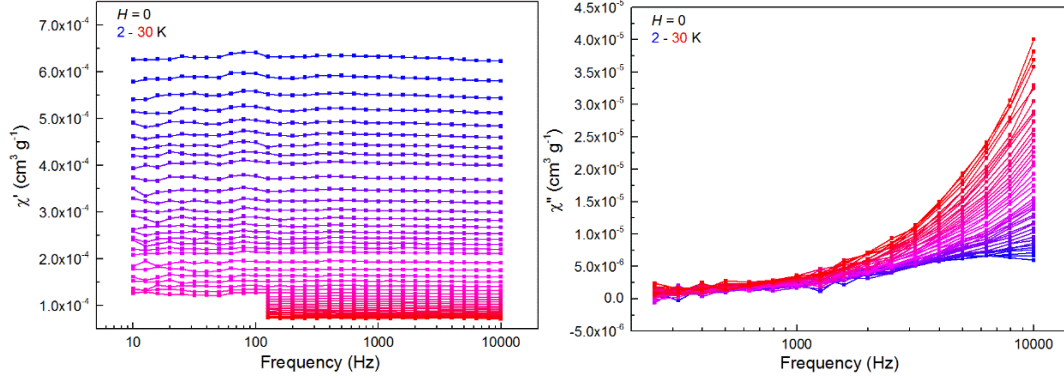
Supplementary Figure 92. Temperature dependence of the $\chi_{\text{mass}}T$ (left) and $\chi_{\text{mole}}T$ (right, normalized by Gd atom number which determined from M vs H data) of $\text{GdCl}_3\text{@SWCNTs}$ in a $H_{\text{DC}} = 1$ kOe applied field.



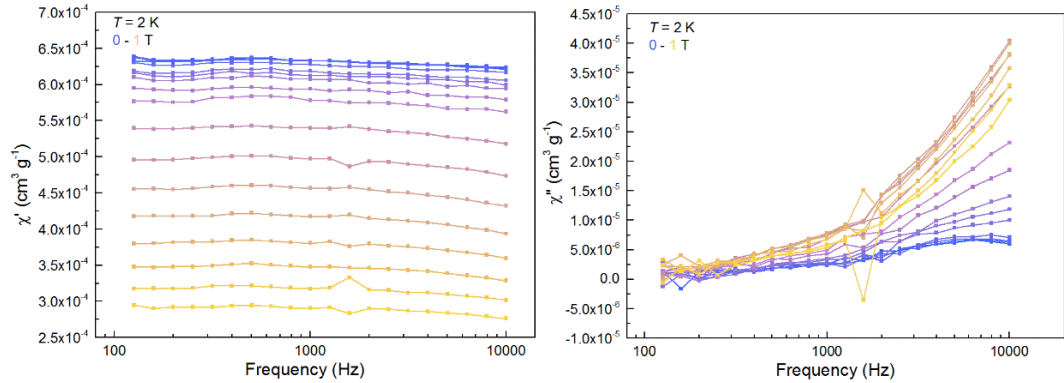
Supplementary Figure 93. Zero-Field cooling (blue) and Field cooling (red) curves of $\text{GdCl}_3\text{@SWCNTs}$ under a 20 Oe applied field.



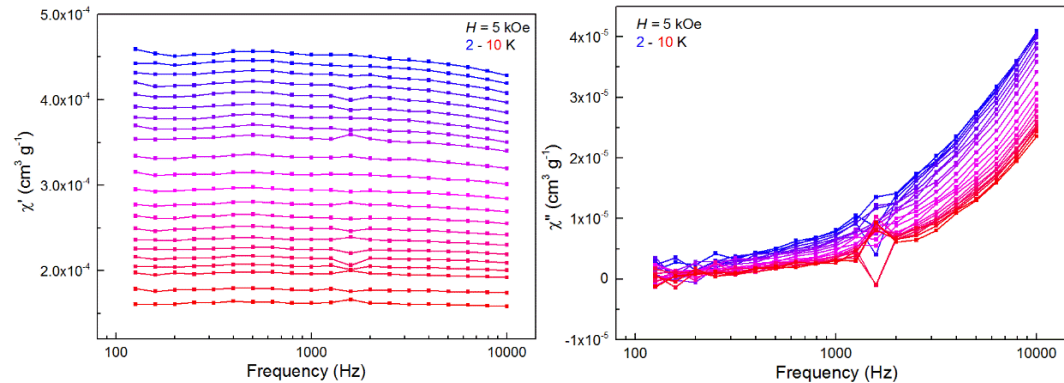
Supplementary Figure 94. Temperature reduced field dependent magnetization for GdCl₃@SWCNTs (left). The mass magnetization comparison between GdCl₃@SWCNTs (black) and GdCl₃ (red) at 5 K (right).



Supplementary Figure 95. The frequency dependent in-phase (χ') (left) and out-of-phase (χ'') (right) mass susceptibility in zero field for GdCl₃@SWCNTs.

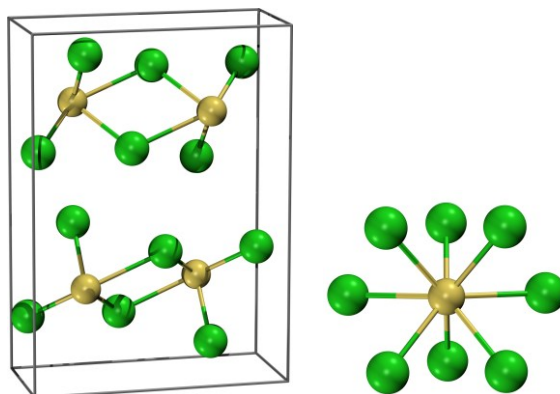


Supplementary Figure 96. The frequency dependent in-phase (χ') (left) and out-of-phase (χ'') (right) mass susceptibility at 2 K under various applied field for GdCl₃@SWCNTs.



Supplementary Figure 97. The frequency dependent in-phase (χ') (left) and out-of-phase (χ'') (right) mass susceptibility in a 5 kOe applied field for GdCl₃@SWCNTs.

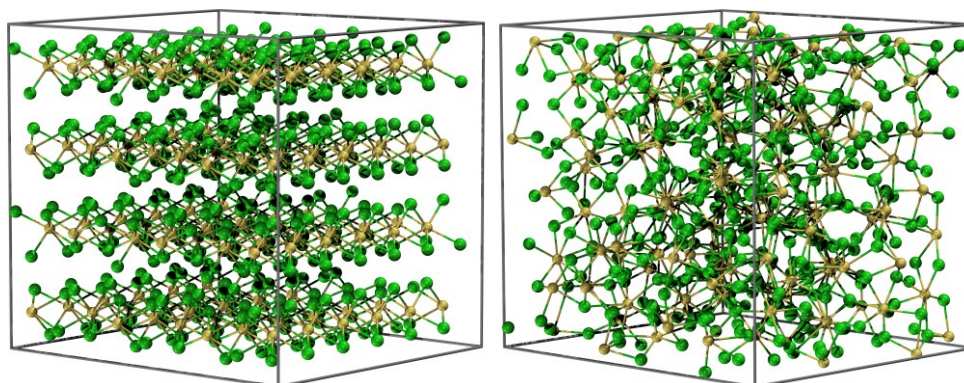
8.4 CASSCF calculation



Supplementary Figure 98. Crystal structure of DyCl₃ (left) and the [DyCl₆]⁵⁻ polyhedral with a quasi-C_{2v} symmetry (right). The colours of the atoms indicate dysprosium (yellow) and chlorine (green).

Supplementary Table 31. The U_{eff} of [DyCl₆]⁵⁻ polyhedral in the crystalline.

System	U_{eff} (10K, cm ⁻¹)	U_{eff} (100K, cm ⁻¹)	U_{eff} (300K, cm ⁻¹)
[DyCl ₆] ⁵⁻ (VDPZ)	16.4	42.3	77.1



Supplementary Figure 99. Ball-and-stick representations of the crystal structure of pristine DyCl₃ (left) and the quenched DyCl₃ structure (right). The colours of the atoms indicate dysprosium (yellow) and chlorine (green).

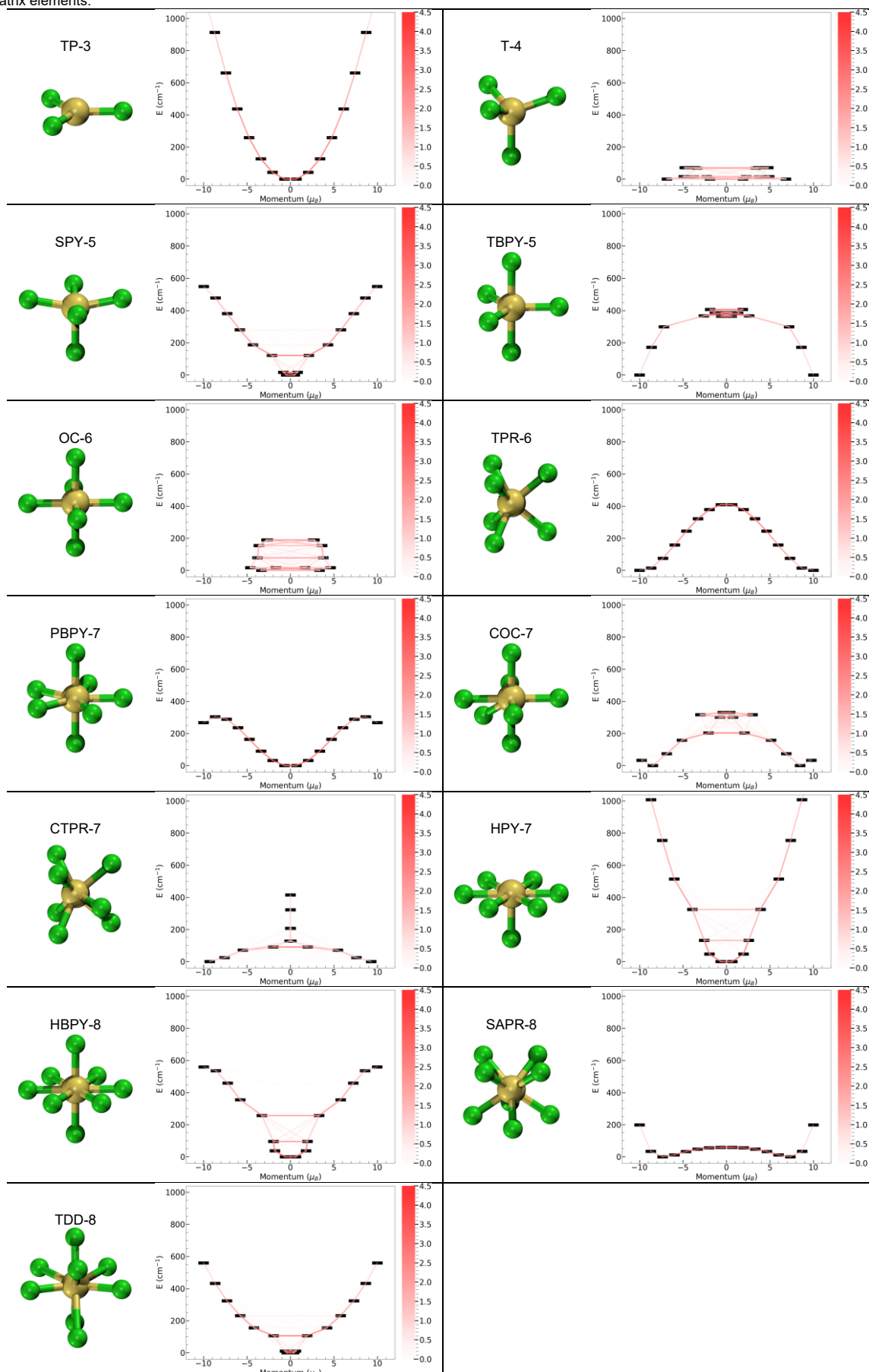
Supplementary Table 32. Statistics of individual polyhedral species in the quenched DyCl₃ structure and their corresponding average U_{eff} .

P.H. type	C.N.	Num.	Avg. U_{eff} (10K, cm ⁻¹)	Avg. U_{eff} (100K, cm ⁻¹)	Avg. U_{eff} (300K, cm ⁻¹)
OC-6	6	10	0.83	119.6	175.6
TPR-6	6	4	0.07	205.1	293.5
COC-7	7	12	0.43	108.8	136.9
PBPY-7	7	16	0.18	117.1	151.1
CTPR-7	7	42	1.10	98.1	125.1
BTPR-8	8	18	2.89	98.7	117.9
JBTPR-8	8	4	6.15	83.1	100.7
SAPR-8	8	17	2.87	85.9	103.4
TDD-8	8	16	2.36	100.9	120.8
CSAPR-9	9	2	4.28	64.9	73.7
MFF-9	9	2	2.11	78.7	93.1
TCTPR-9	9	1	0.13	123.4	138.4
Sum	7.35	144	1.66	103.6	131.5

Supplementary Table 33 Main parameters of the selected representative structures.

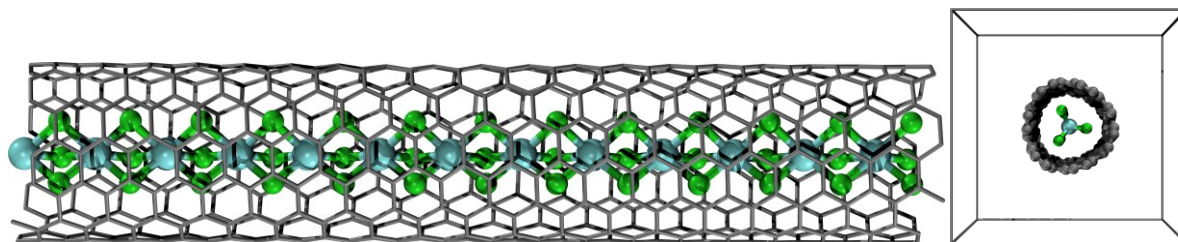
model	No.	Num.	diameter (Å)	c (Å)	$\bar{E}$ (eV)	Avg. U_{eff} (10K, cm ⁻¹)	Avg. U_{eff} (100K, cm ⁻¹)	Avg. U_{eff} (300K, cm ⁻¹)	Polyhedron
TX	214	6	10.68	8.02	-1.946	1.2	75.6	128.6	(OC-6)×6
TZ	001	4	7.81	7.26	-2.369	0.9	74.5	132.0	(CTPR-7)×4
TS	217	6	8.79	12.18	-2.044	1.9	141.1	188.5	(OC-6)×4,(TPR-6)×2
TB	054	8	10.51	13.56	-1.948	2.2	109.0	149.8	(OC-6)×7,(SAPR-8)×1
TH	108	16	9.36	27.19	-2.212	6.7	276.7	383.7	(TPR-6)×16
TU	186	5	9.09	9.15	-1.973	1.3	69.3	100.5	(SPY-5)×1,(OC-6)×4

Supplementary Table 34. Ideal $[\text{DyCl}_n]^{3-n}$ polyhedra with Dy–Cl bond lengths of 2.785 Å and their corresponding Zeeman eigenstates (black bars) along with transition matrix elements.



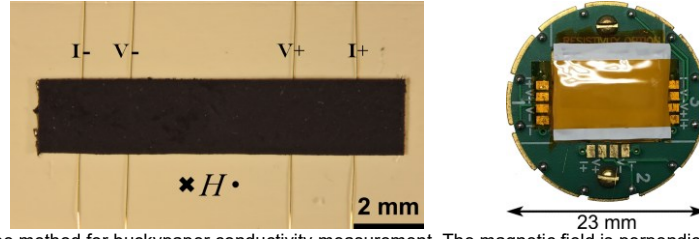
Supplementary Table 35. The U_{eff} of ideal $[\text{DyCl}_n]^{3-n}$ polyhedra with Dy–Cl bond lengths of 2.785 Å.

[DyCl _n] ³⁻ⁿ polyhedron	U_{eff} (cm ⁻¹)		
	(10K)	(100K)	(300K)
TP-3	0.00	2.35	11.84
T-4	2.08	20.43	27.77
SPY-5	1.17	17.20	40.04
TBPY-5	0.01	377.28	383.06
OC-6	1.95	31.10	65.75
TPR-6	13.85	385.78	402.26
PBPY-7	0.00	0.01	0.04
COC-7	0.00	185.26	257.90
CTPR-7	3.86	101.20	162.70
HPY-7	0.00	11.84	55.42
HBPY-8	0.00	18.05	61.03
SAPR-8	24.99	57.19	57.31
TDD-8	1.69	16.47	36.53

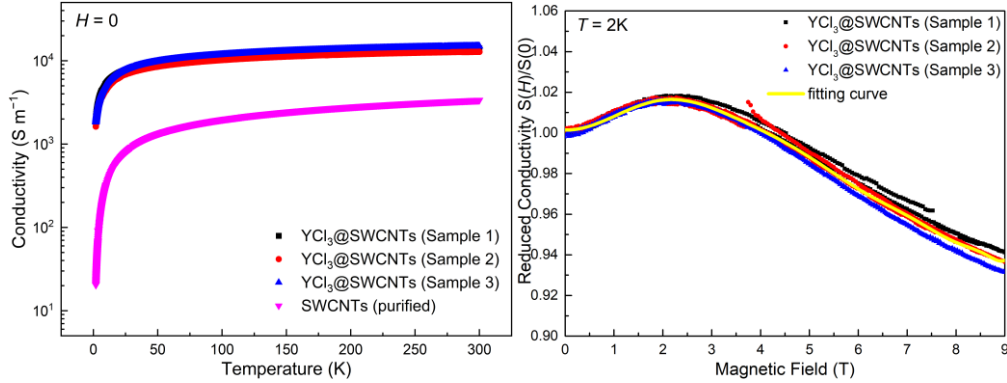


Supplementary Figure 100. The model of a hypothetical $\{\text{YCl}_3\}_{13}@\text{SWCNT}(7,6)$, involving trigonal prism YCl_6 polyhedra. The diameter of $\text{SWCNT}(7,6)$ is 8.85 Å.

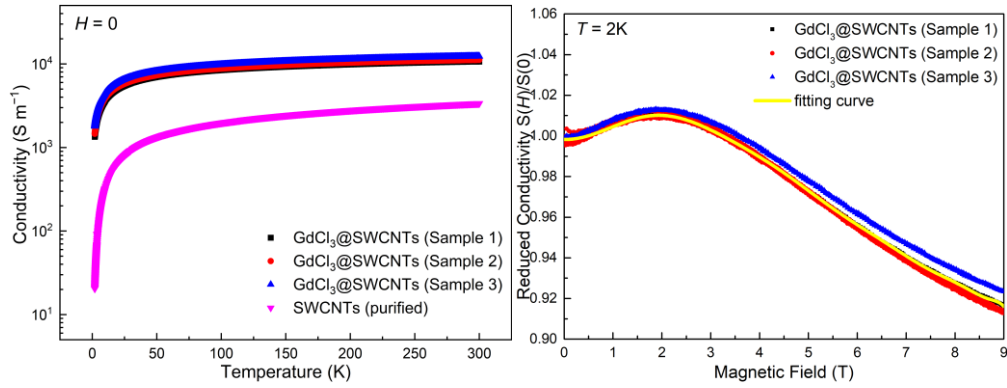
9 Conductivity



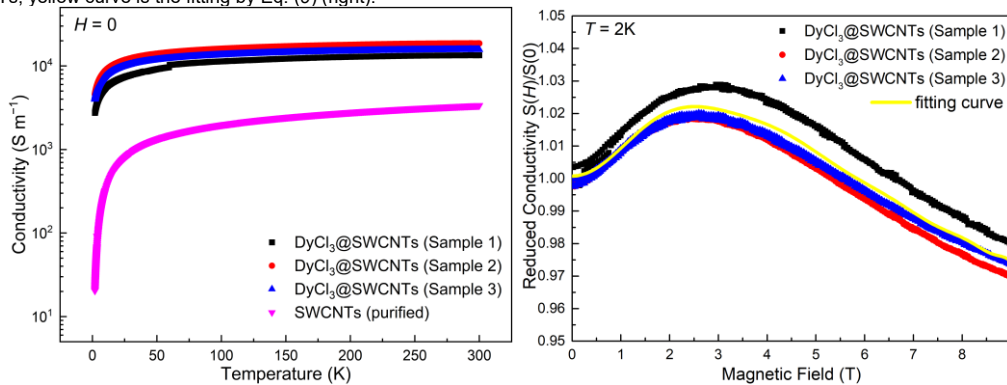
Supplementary Figure 101. Four probe method for buckypaper conductivity measurement. The magnetic field is perpendicular to the film (left). The assembled sample stage (right).



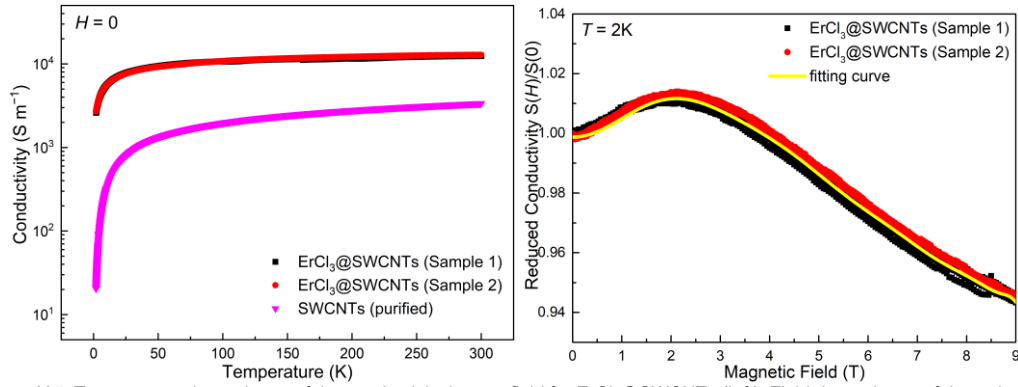
Supplementary Figure 102. Temperature dependence of the conductivity in zero field for YCl₃@SWCNTs (left). Field dependence of the reduced conductivity at 2 K for YCl₃@SWCNTs, yellow curve is the fitting by Eq. (9) (right).



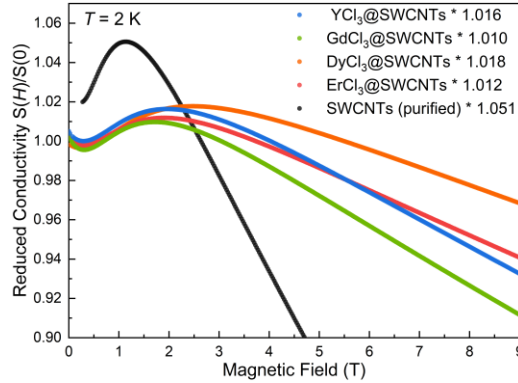
Supplementary Figure 103. Temperature dependence of the conductivity in zero field for GdCl₃@SWCNTs (left). Field dependence of the reduced conductivity at 2 K for GdCl₃@SWCNTs, yellow curve is the fitting by Eq. (9) (right).



Supplementary Figure 104. Temperature dependence of the conductivity in zero field for DyCl₃@SWCNTs (left). Field dependence of the reduced conductivity at 2 K for DyCl₃@SWCNTs, yellow curve is the fitting by Eq. (9) (right).



Supplementary Figure 105. Temperature dependence of the conductivity in zero field for ErCl_3 @SWCNTs (left). Field dependence of the reduced conductivity at 2 K for ErCl_3 @SWCNTs, yellow curve is the fitting by Eq. (9) (right).



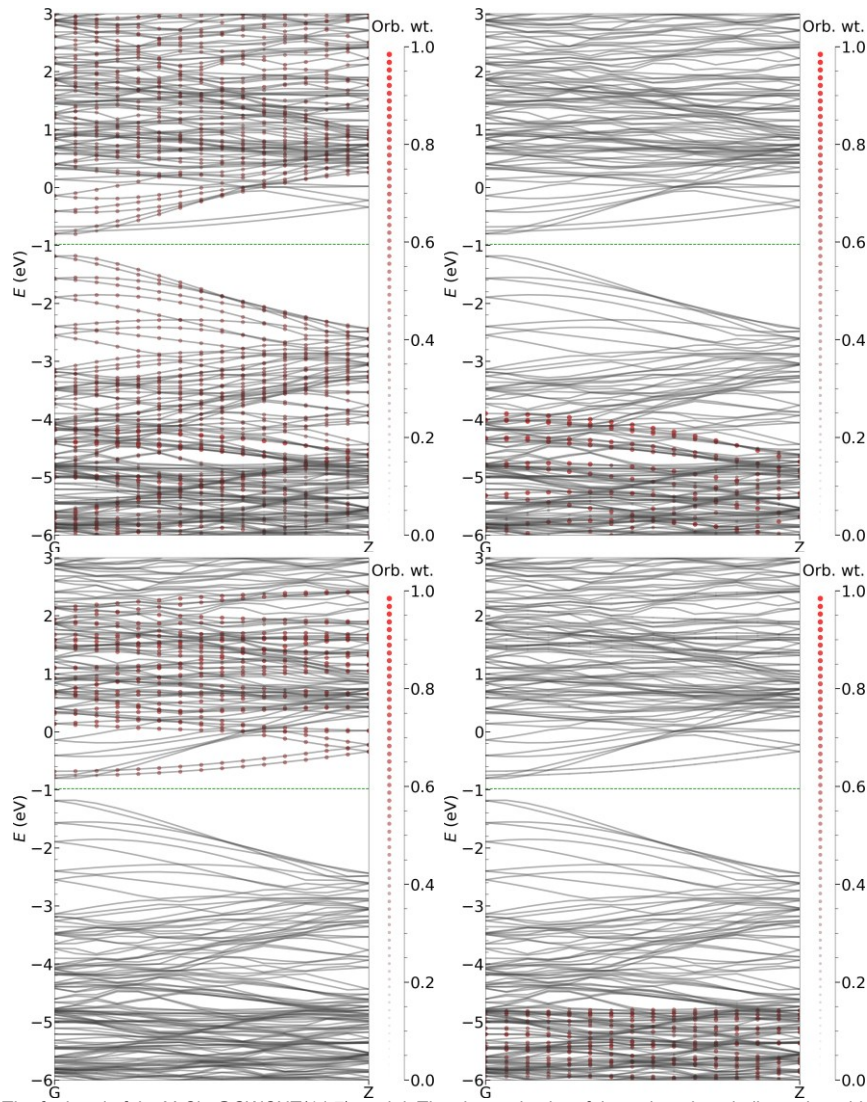
Supplementary Figure 106. The fitting curves of the field dependence of the reduced conductivity at 2 K for purified 1.4nm SWCNTs (black), YCl_3 @SWCNTs (green), GdCl_3 @SWCNTs (magenta), DyCl_3 @SWCNTs (red), and ErCl_3 @SWCNTs (blue).

Supplementary Table 36. The fitting parameter from the field dependence of the reduced conductivity at 2 K for purified 1.4nm SWCNTs, DyCl_3 @SWCNTs, ErCl_3 @SWCNTs, GdCl_3 @SWCNTs, and YCl_3 @SWCNTs. Multiple samples were fitted simultaneously using Eq. (9). The first sample was treated as the reference, and each of the remaining samples was fitted using Eq. (9) multiplied by an additional overall scaling factor (C_2 , C_3).

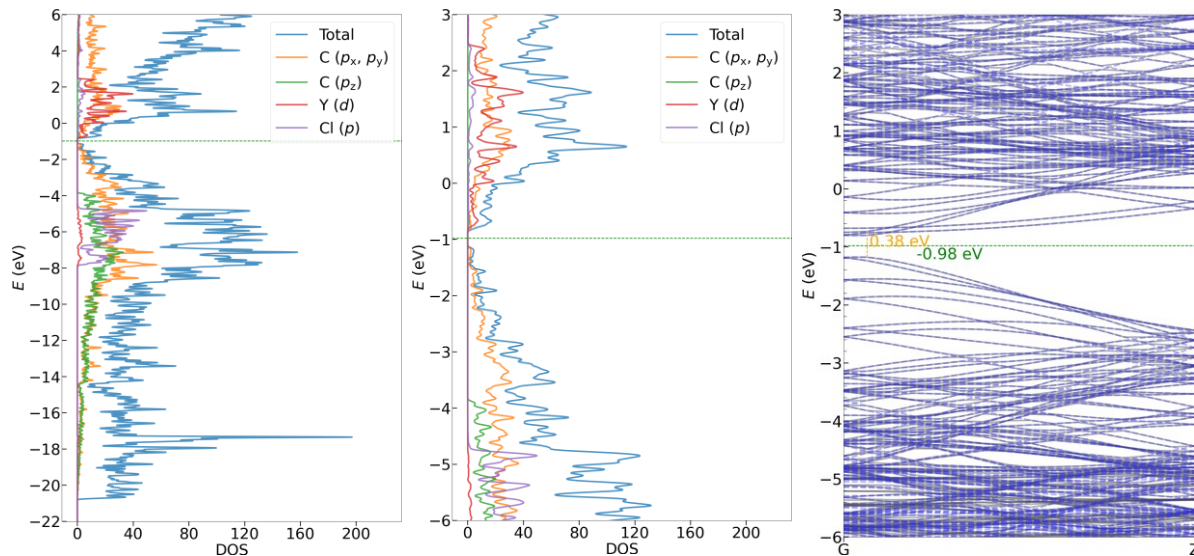
	SWCNTs (purified)	YCl_3 @SWCNTs	GdCl_3 @SWCNTs	DyCl_3 @SWCNTs	ErCl_3 @SWCNTs
a	-1.47E-01	-2.57E-02	-3.40E-02	-1.48E-02	-2.28E-02
a (error)	4.19E-02	3.62E-03	2.49E-03	2.84E-03	1.92E-03
b	4.16E-01	2.45E-01	2.31E-01	2.19E-01	2.19E-01
b (error)	--	5.17E+02	2.39E+02	4.78E+02	--
c	2.27E+00	7.11E-01	8.89E-01	5.87E-01	7.67E-01
c (error)	--	3.90E+02	2.60E+02	3.20E+02	--
d	1.46E+00	5.26E-01	6.38E-01	4.41E-01	5.79E-01
d (error)	1.60E-01	8.40E-03	6.19E-03	8.92E-03	6.40E-03
e	6.37E-01	7.63E-01	7.73E-01	7.90E-01	7.84E-01
e (error)	--	5.17E+02	2.39E+02	4.78E+02	--
m	6.14E-01	8.21E-01	7.60E-01	8.72E-01	7.97E-01
m (error)	7.55E-02	3.99E-02	2.03E-02	5.54E-02	2.39E-02
n	1.85E+00	1.87E+00	1.84E+00	1.92E+00	1.91E+00
n (error)	3.07E-01	9.24E-02	5.45E-02	9.80E-02	6.03E-02
C_2	1.01E+00	9.97E-01	9.99E-01	9.90E-01	1.00E+00
C_2 (error)	1.72E-03	1.29E-04	8.89E-05	1.15E-04	7.75E-05
C_3	9.60E-01	9.94E-01	1.00E+00	9.92E-01	1.00E+00
C_3 (error)	1.68E-03	1.29E-04	8.92E-05	1.15E-04	7.75E-05
Reduced Chi-Sqr	4.34E-04	5.86E-06	4.06E-06	2.46E-06	2.63E-06
Adj. R-Square	9.55E-01	9.92E-01	9.96E-01	9.90E-01	9.95E-01

10 Electronic band structure and charge transport properties

10.1 The $\text{Y}_6\text{Cl}_{18}\text{@SWCNT}(14,7)$ model



Supplementary Figure 107. The fat band of the $\text{Y}_6\text{Cl}_{18}\text{@SWCNT}(14,7)$ model. The size and color of the red markers indicate the orbital contributions to each energy band: carbon p_x and p_y orbitals (top left), carbon p_z orbital (top right), yttrium d orbitals (bottom left), and chlorine p orbitals (bottom right).

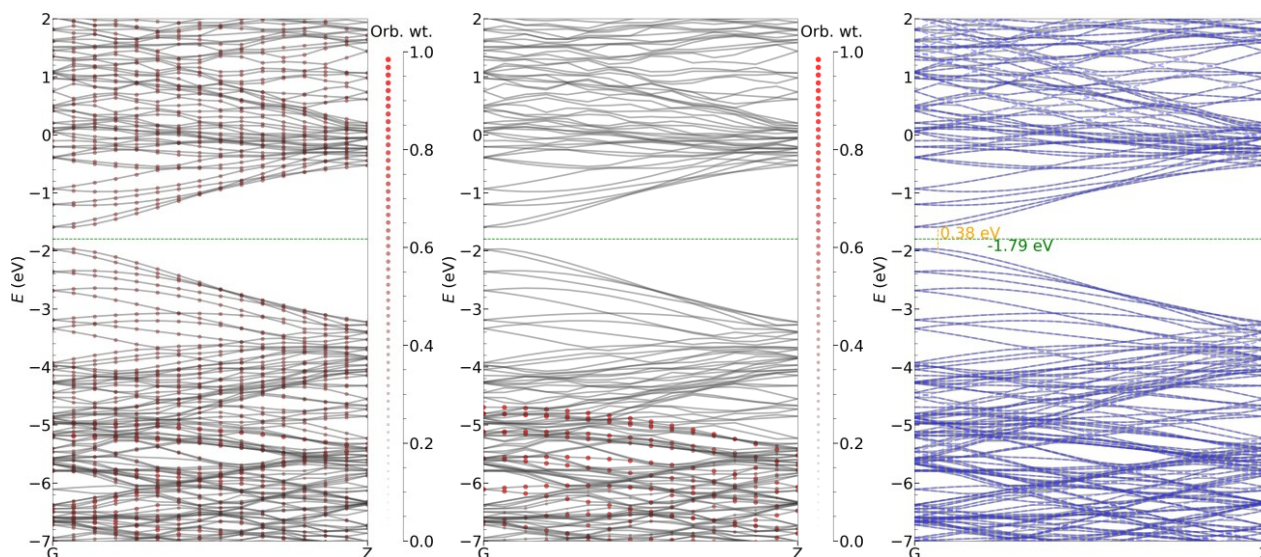


Supplementary Figure 108. The DOS and PDOS of the $\text{Y}_6\text{Cl}_{18}\text{@SWCNT}(14,7)$ model are shown on the left, with an enlarged view near the Fermi level in the center. On the right, the electronic band structure is presented: grey solid lines represent DFT-calculated bands, and blue dashed lines represent bands obtained via MLWFs interpolation. The system has a band gap of 0.38 eV.

Supplementary Table 37. The Hirshfeld-I charge of the $\text{Y}_6\text{Cl}_{18}@\text{SWCNT}(14,7)$ model.

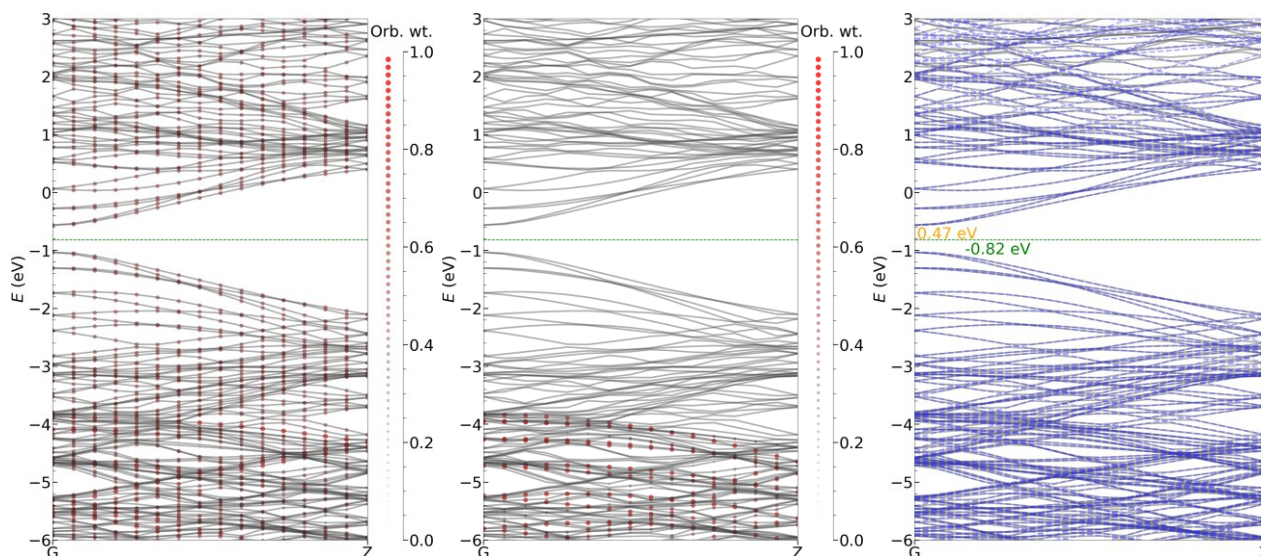
Elements	Number	Hirshfeld-I
C	196	-0.001
Y	6	1.567
Cl	18	-0.512

10.2 The deformed SWCNT(14,7) model



Supplementary Figure 109. The fat band of the deformed SWCNT(14,7) model. The size and color of the red markers indicate the orbital contributions to each energy band: carbon p_x and p_y orbitals (left), carbon p_z orbital (middle). The electronic band structure is presented: grey solid lines represent DFT-calculated bands, and blue dashed lines represent bands obtained *via* MLWFs interpolation. The system has a band gap of 0.38 eV (right).

10.3 The pristine SWCNT(14,7) model.



Supplementary Figure 110. The fat band of the pristine SWCNT(14,7) model. The size and color of the red markers indicate the orbital contributions to each energy band: carbon p_x and p_y orbitals (left), carbon p_z orbital (middle). The electronic band structure is presented: grey solid lines represent DFT-calculated bands, and blue dashed lines represent bands obtained *via* MLWFs interpolation. The system has a band gap of 0.47 eV (right).