Supporting Information

Ab Initio Structure Determination of $Cu_{2-x}Te$ Plasmonic Nanocrystals by Precession-Assisted Electron Diffraction Tomography and HAADF-STEM Imaging

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Table S1. EDS measurements performed on the same NSs used for EDT data collections. Measurements were done on 13 single NSs by a Bruker EDS detector XFlash6T-60 mounted in a Zeiss Libra120 TEM.

Spectrum	**Cu-K (at. %)**	**Te-L (at. %)**
NS1	56.80	43.2
NS2	54.32	45.68
NS3	63.83	36.17
NS4	58.21	41.79
NS5	57.17	42.83
NS6	60.07	39.93
NS7	58.15	41.85
NS8	58.40	41.60
NS9	58.71	41.29
NS10	54.47	45.53
NS11	61.72	38.28
NS12	56.54	43.46
NS13	56.87	43.13
Average	58.25	41.75
Standard deviation	2.38	2.38

Table S2. Structural and experimental details regarding the structure solution of $Cu_{1.5}Te$ phase. *: cell parameters refined by Rietveld refinement on XRPD data. **: agreement parameters are rather high for this structure, even for EDT standard. This can be explained with the fact that at least part of the Cu positions determined are in fact partially occupied and there must by other low occupied sites assigned as vacancies. Moreover, surface effects, twinning and stacking disorder are expected to significantly deteriorate the agreement parameters for this sample.

Crystallographic details	
Unit cell content (as determined by EDT)	$Cu_{124}Te_{96}$
Space group	$P222_1$
a, Å	7.544(3)*
b, Å	22.622(7)*
c, Å	29.797(4)*
Cell volume, Å^3	5085(3)*
EDT data collection	
Cell volume, Å^3	5060
Tilt range, °	130
Tilt step, °	1
Precession semi-angle, °	1
***Ab initio* structure determination (SIR2014)**	
Data resolution, Å	0.9
N° sampled reflections	12707
N° independent reflections	3192
Independent reflection coverage, %	77
R_{sym}, %	27.04
R_{SIR}, %	34.61**

Least-squares refinement (SHELXL)	
Data resolution, Å	0.9
***N*° *reflections* > 4σ**	3702
R_{int}, %	36.82
***R*1 (4σ), %**	46.55**
GooF	2.660**

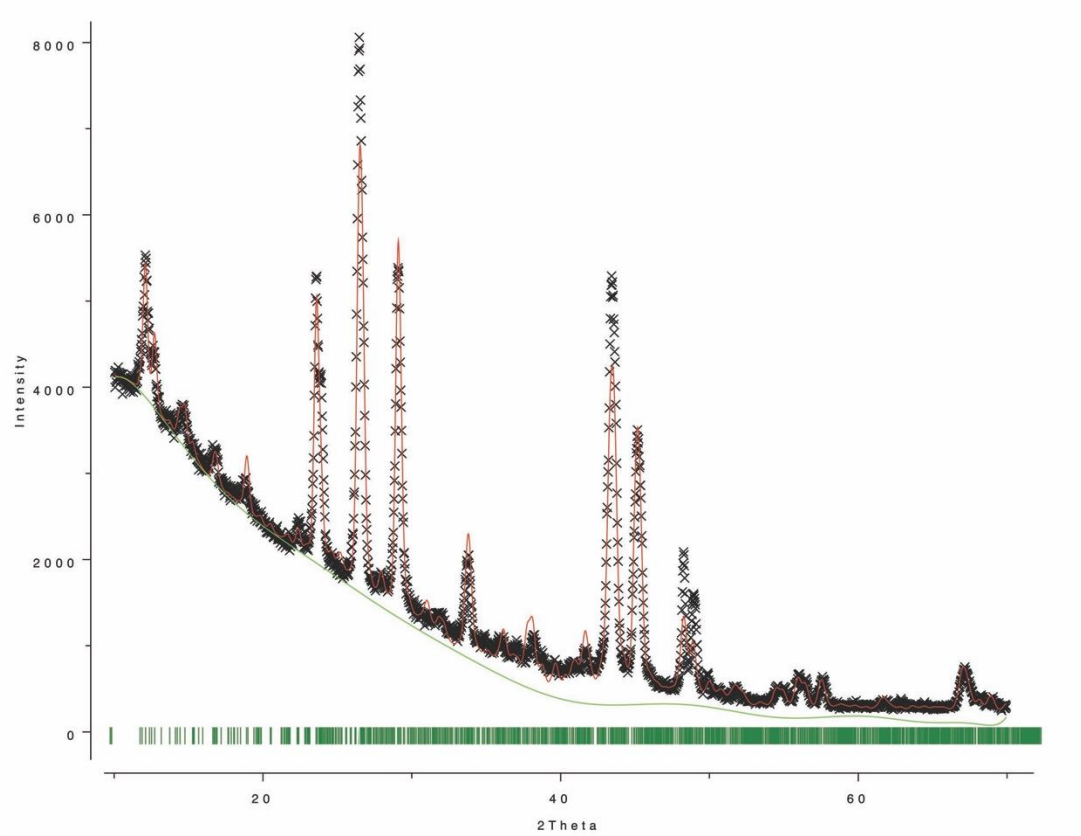

Figure S1. Rietveld fit obtained with the structure solved with EDT data. Only the unit cell, the profile and background parameters have been refined.

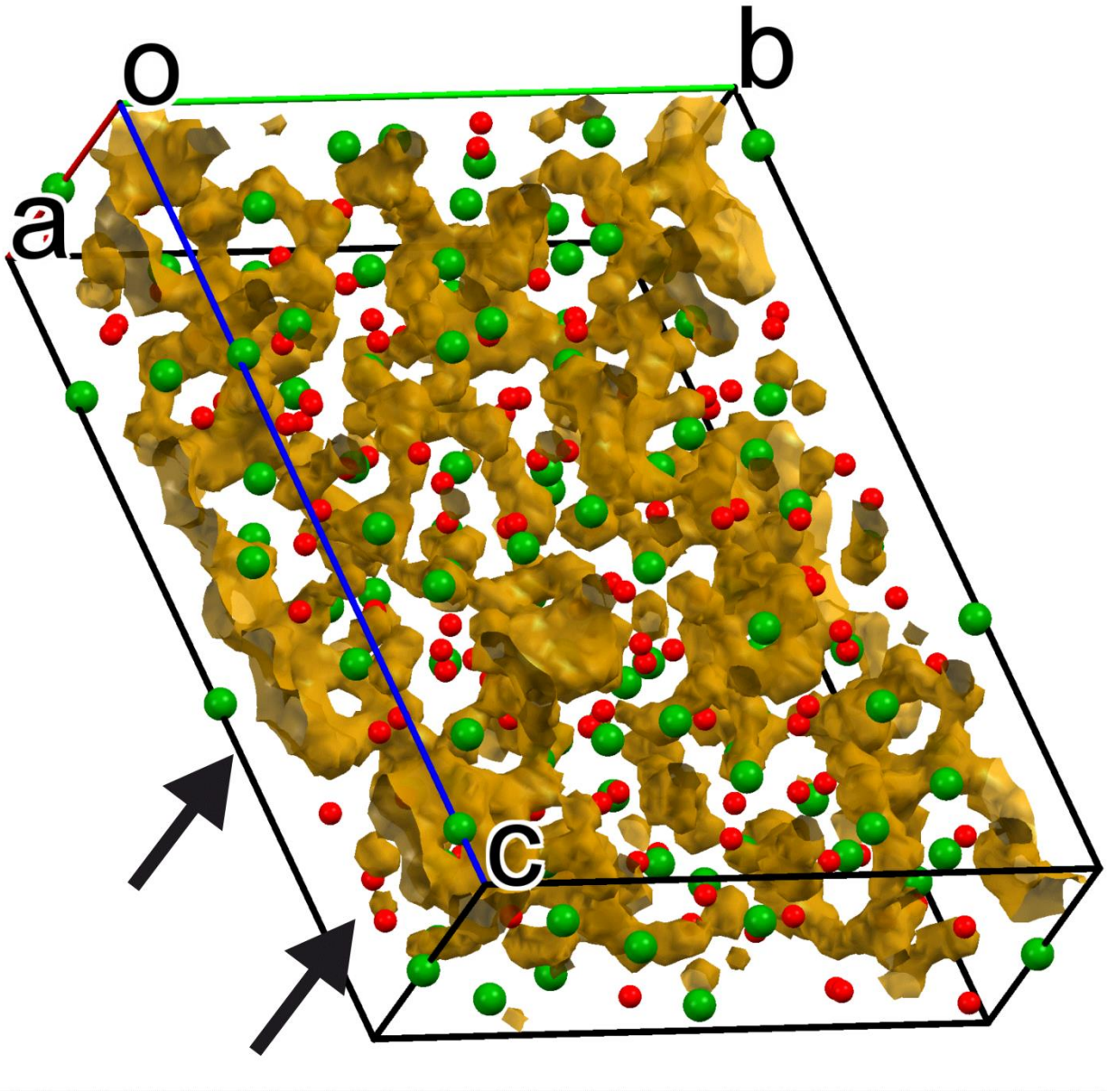

Figure S2. Low-density areas in $Cu_{1.5}Te$ structure. The arrow marks one of the low-density areas parallel to (100) and responsible for the up-up-down-down pattern in [010] HAADF-STEM images.

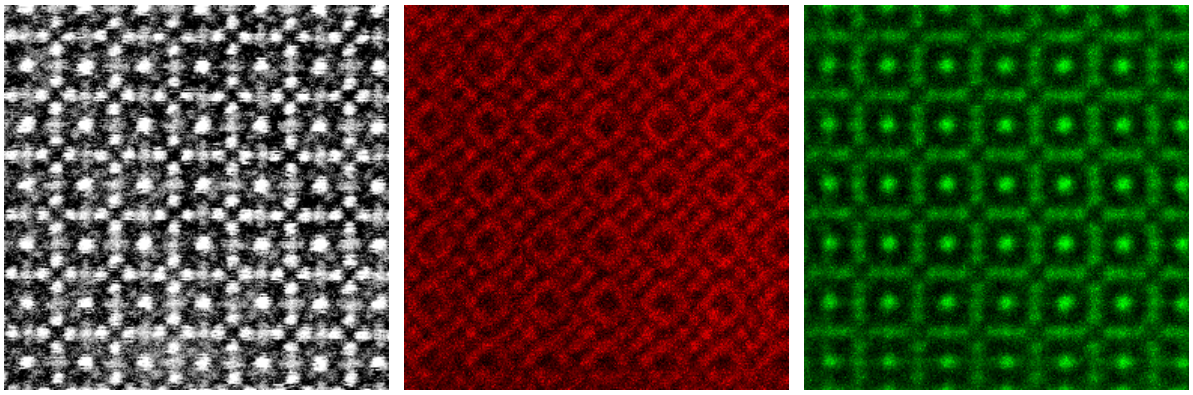

Figure S3. ADF image, together with the Cu-K EDS map (red) and Te-L EDS map (green)