

**Supporting information for**

**Nanotubes from the misfit compound alloy LaS-Nb<sub>x</sub>Ta<sub>(1-x)</sub>S<sub>2</sub>**

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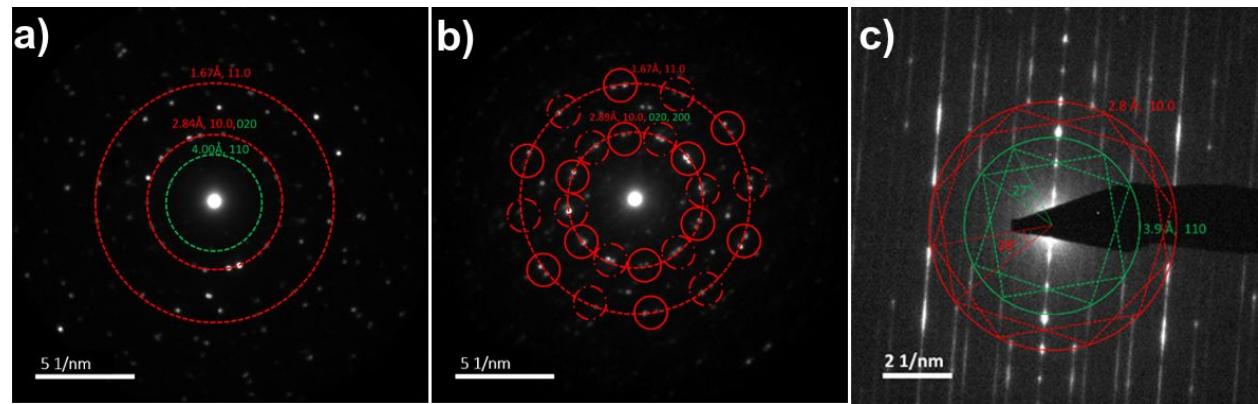
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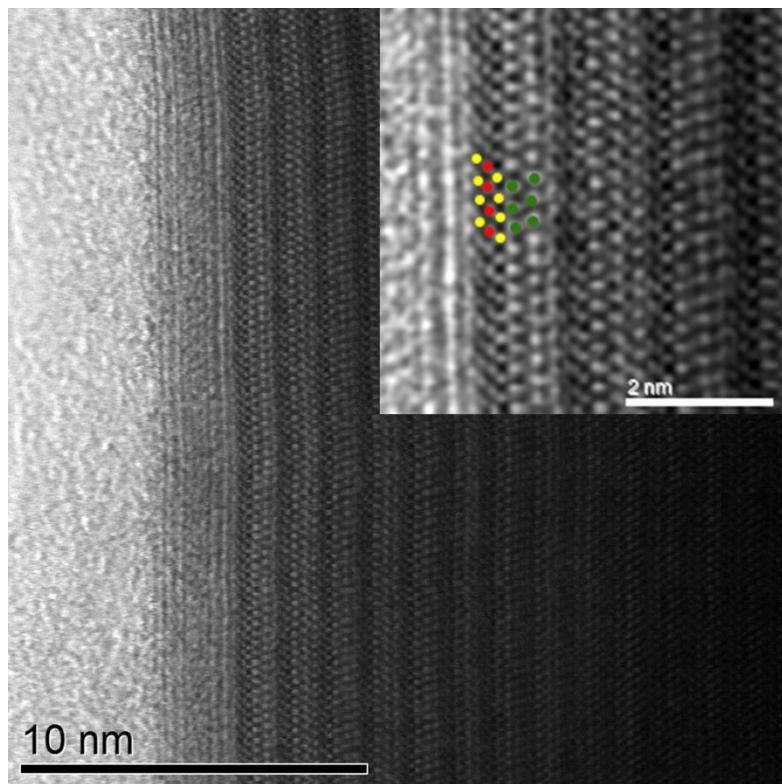
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**Fig. S1.** ED in STEM mode of a. nanotube prepared from 20 %at Nb, b. 40 at% Nb, c. 80 at %. the ED of a 80 at% Nb tube. The green and red circles are overlaid on the reflections from the (110) planes of the LaS layers and the (100) planes of the  $\text{Nb}_x\text{Ta}_{(1-x)}\text{S}_2$  slabs of the nanotube, respectively.



**Fig. S2.** HR-STEM BF of the nanotube shown in **Fig. 6** (prepared from 80 at% Nb concentration). The inset shows a magnified view of the filtered image superposed with the atomic model (green: La, red : Ta/Nb, yellow : S). Note the 1T (octahedral) structure of the hexagonal  $\text{LaS-Nb}_x\text{Ta}_{(1-x)}\text{S}_2$  lattice.

NT normal 1	Center (180 nm from surface)	NT normal 1	90 nm from surface
Element	Atomic Fraction (%)	Element	Atomic Fraction (%)
S	65.78	S	63.24
Nb	8.40	Nb	8.14
La	17.04	La	18.36
Ta	8.78	Ta	10.26
Nb/Ta	0.96	Nb/Ta	0.79
La/(Nb+Ta)	0.99	La/(Nb+Ta)	1.00
Nb+Ta for 1.15 La	1.16	Nb+Ta for 1.15 La	1.15

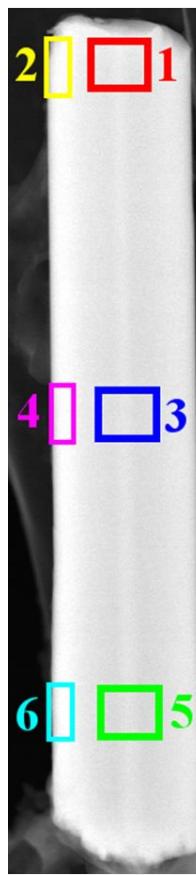
NT normal 1	30 nm from surface
Element	Atomic Fraction (%)
S	68.39
Nb	6.69
La	15.25
Ta	9.67
Nb/Ta	0.69
La/(Nb+Ta)	0.93
Nb+Ta for 1.15 La	1.23

**Table S1.** Results of the EDS quantification performed in various areas of the nanotube with a normal periodicity (shown in **Fig. 6a**).



NT normal 1	Extreme surface
Element	Atomic Fraction (%)
S	17.22
Nb	1.46
La	60.11
Ta	21.20
Nb/Ta	0.07
La/(Nb+Ta)	2.65
Nb+Ta for 1.15 La	0.43

**Fig. S3.** *Top:* HR-STEM HAADF of the NT shown in Fig. 6a. *Bottom:* Results of the EDS quantification performed at the area highlighted by the red square.



The figure shows a scanning electron micrograph (SEM) of a nanotube. Six measurement points are indicated by numbered boxes: 1 (red), 2 (red), 3 (blue), 4 (blue), 5 (red), and 6 (blue). The table below provides EDS quantification results for these areas.

Area 1		Area 2	
Element	Atomic Fraction (%)	Element	Atomic Fraction (%)
S	60.80	S	54.80
Nb	17.09	Nb	19.80
La	17.09	La	18.97
Ta	5.02	Ta	6.43
Nb/Ta	3.40	Nb/Ta	3.08
La/(Nb+Ta)	0.77	La/(Nb+Ta)	0.72
Nb+Ta for 1.15 La	1.49	Nb+Ta for 1.15 La	1.59

Area 3		Area 4	
Element	Atomic Fraction (%)	Element	Atomic Fraction (%)
S	61.67	S	61.67
Nb	10.90	Nb	10.90
La	17.53	La	17.53
Ta	9.90	Ta	9.90
Nb/Ta	1.10	Nb/Ta	1.31
La/(Nb+Ta)	0.84	La/(Nb+Ta)	1.00
Nb+Ta for 1.15 La	1.36	Nb+Ta for 1.15 La	1.15

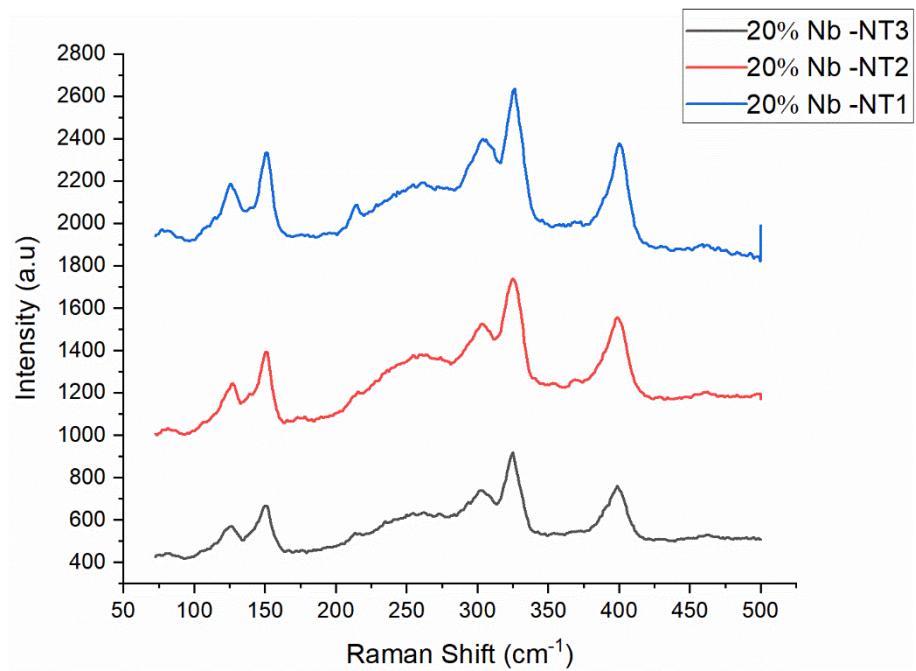
  

Area 5		Area 6	
Element	Atomic Fraction (%)	Element	Atomic Fraction (%)
S	63.41	S	53.09
Nb	7.92	Nb	15.18
La	17.79	La	19.79
Ta	10.88	Ta	11.94
Nb/Ta	0.73	Nb/Ta	1.27
La/(Nb+Ta)	0.95	La/(Nb+Ta)	0.73
Nb+Ta for 1.15 La	1.22	Nb+Ta for 1.15 La	1.58

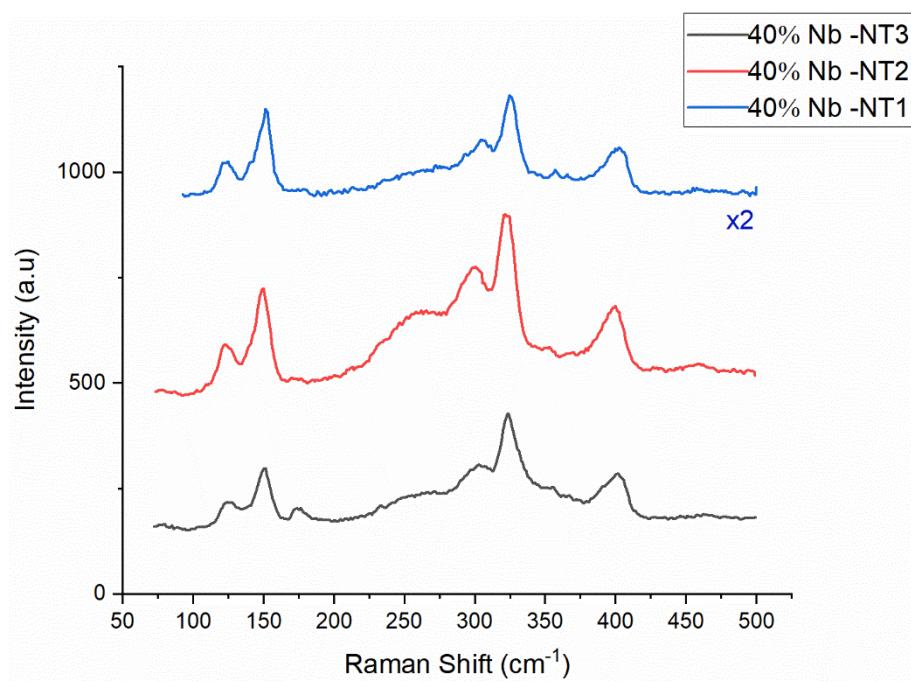
**Fig. S4.** Results of the EDS quantification performed in various areas of the nanotube showing a juxtaposition of normal and double periodicity.

NT double 1			NT double 2	
Element	Atomic Fraction (%)		Element	Atomic Fraction (%)
S	56.41		S	53.34
Nb	17.5		Nb	15.06
La	20.12		La	21.66
Ta	5.97		Ta	9.93
Nb/Ta	2.93		Nb/Ta	1.52
La/(Nb+Ta)	0.86		La/(Nb+Ta)	0.87
Nb+Ta for 1.15 L	1.34		Nb+Ta for 1.15 La	1.33

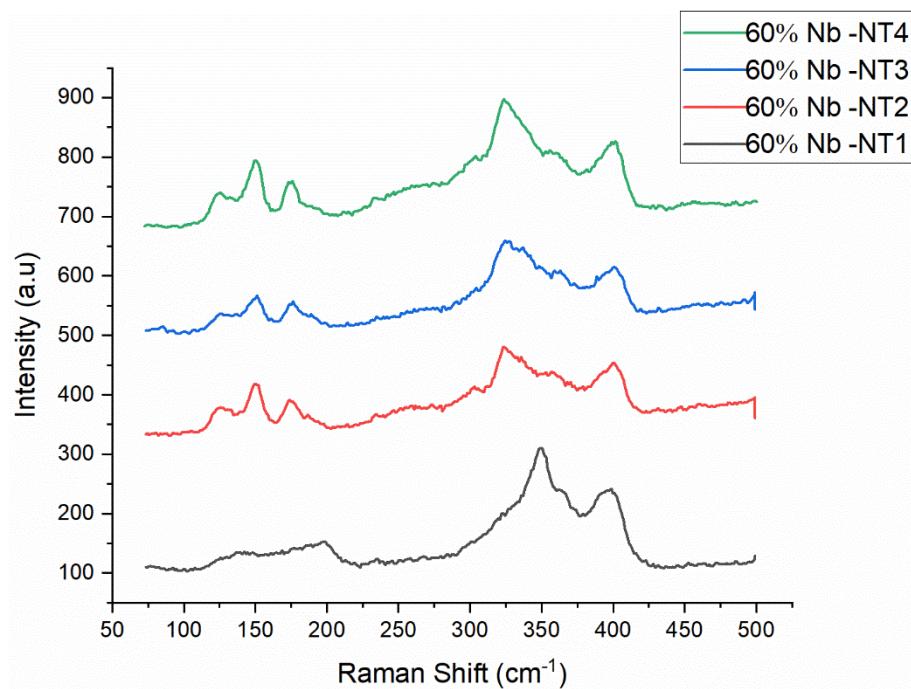
**Table S2.** Results of the EDS quantification performed on two different nanotubes showing a double periodicity.



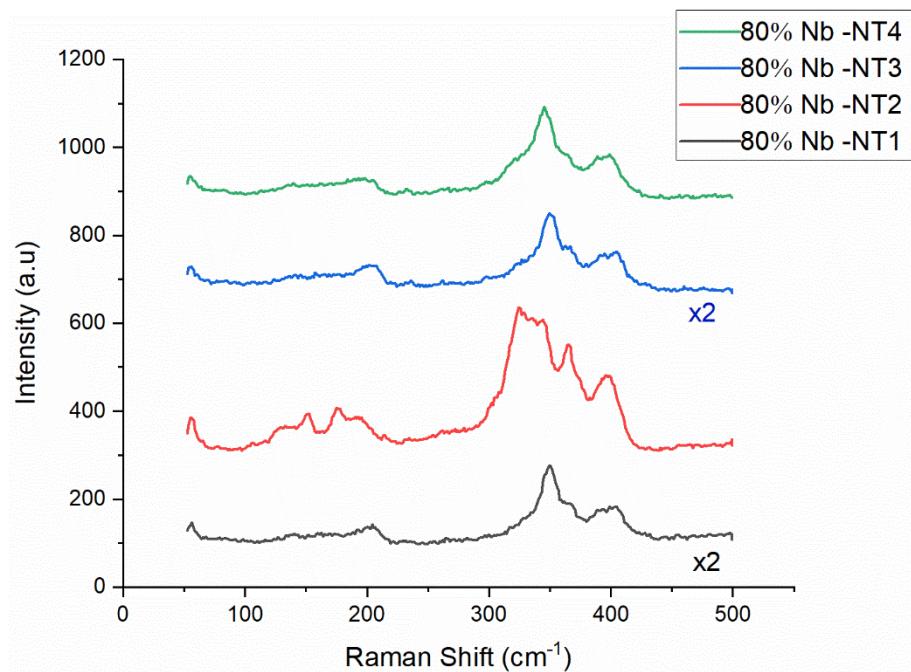
**Fig. S5.** Raman spectra of a few MLC LaS-Nb<sub>x</sub>Ta<sub>(1-x)</sub>S<sub>2</sub> nanotubes prepared from a precursor containing 20 at% niobium (and 80 at% tantalum). The spectra does not vary much from one tube to the other.



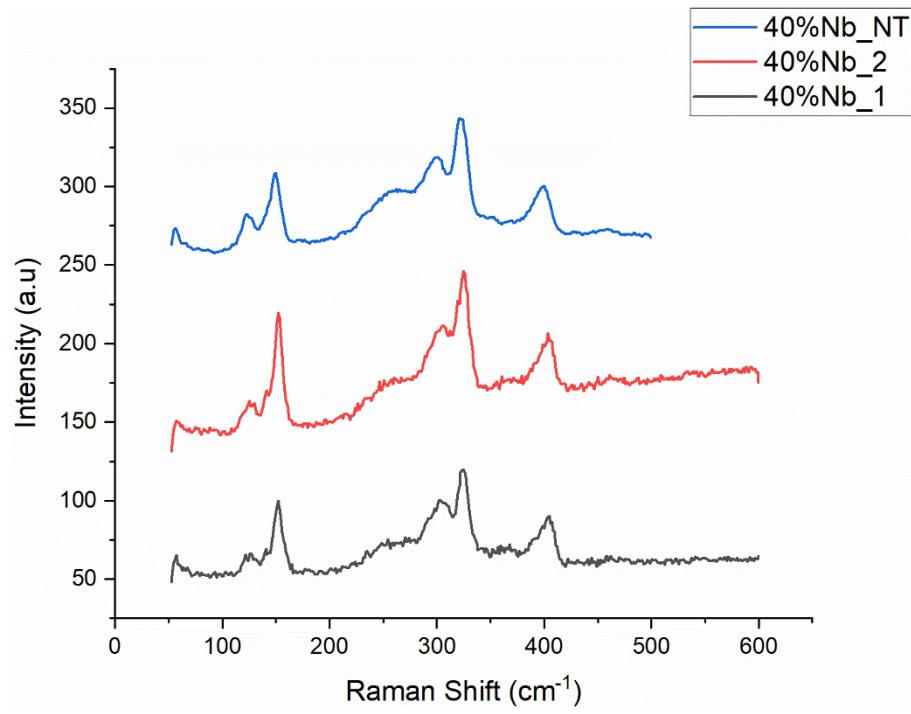
**Fig. S6.** Raman spectra of a few MLC LaS-Nb<sub>x</sub>Ta<sub>(1-x)</sub>S<sub>2</sub> nanotubes prepared from a precursor containing 40 at% niobium (and 60 at% tantalum). All spectra look quite similar.



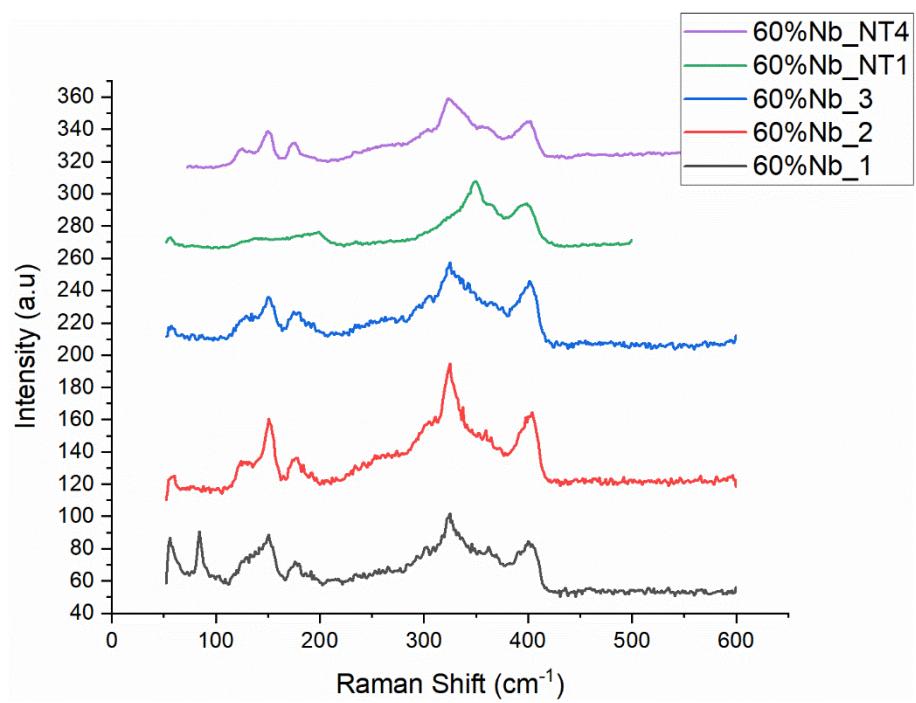
**Fig. S7.** Raman spectra of a few MLC LaS-Nb<sub>x</sub>Ta<sub>(1-x)</sub>S<sub>2</sub> nanotubes prepared from a precursor containing 60 at% niobium (and 40 at% tantalum).



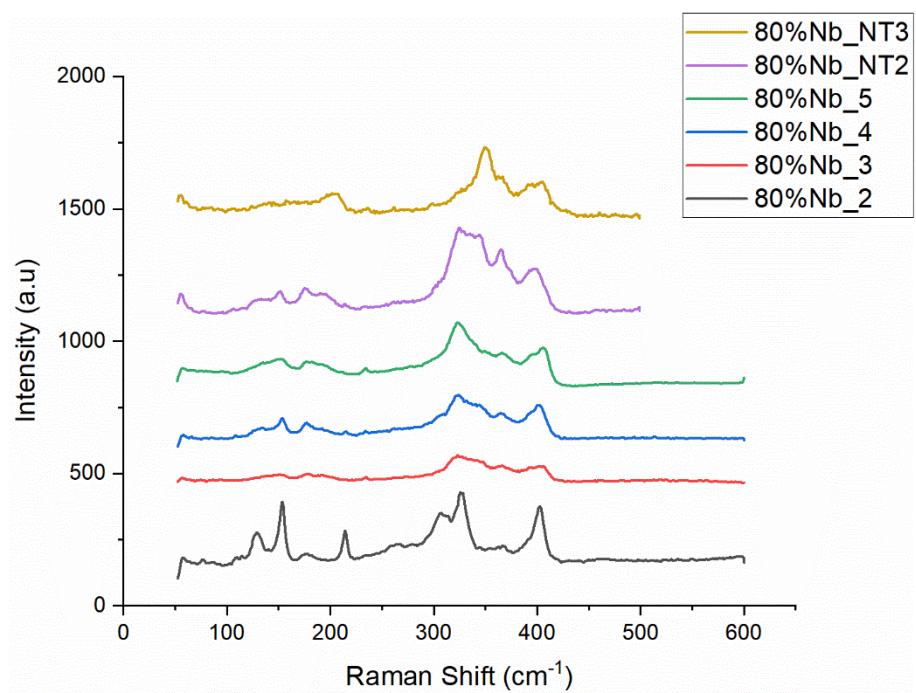
**Fig. S8.** Raman spectra of a few MLC LaS-Nb<sub>x</sub>Ta<sub>(1-x)</sub>S<sub>2</sub> nanotubes prepared from a precursor containing 80 at% niobium (and 20 at% tantalum)



**Fig. S9.** Raman spectra of a few MLC LaS-Nb<sub>x</sub>Ta<sub>(1-x)</sub>S<sub>2</sub> platelets prepared from a precursor containing 40 at% niobium (and 60 at% tantalum). Note the similarity to the spectra in **Fig. S6**. However, the peaks are much narrower in the case of the platelets compared to the tubes, which is indicative of the higher crystalline order of the platelets.



**Fig. S10.** Raman spectra of a few MLC LaS-Nb<sub>x</sub>Ta<sub>(1-x)</sub>S<sub>2</sub> platelets prepared from a precursor containing 60 at% niobium (and 40 at% tantalum). The peak in 175 cm<sup>-1</sup> is typical for the tubes (platelets) with high Nb content.



**Fig. S11.** Raman spectra of a few MLC LaS-Nb<sub>x</sub>Ta<sub>(1-x)</sub>S<sub>2</sub> platelets prepared from a precursor containing 80 at% niobium (and 20 at% tantalum).