Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Electronic Supplementary Information

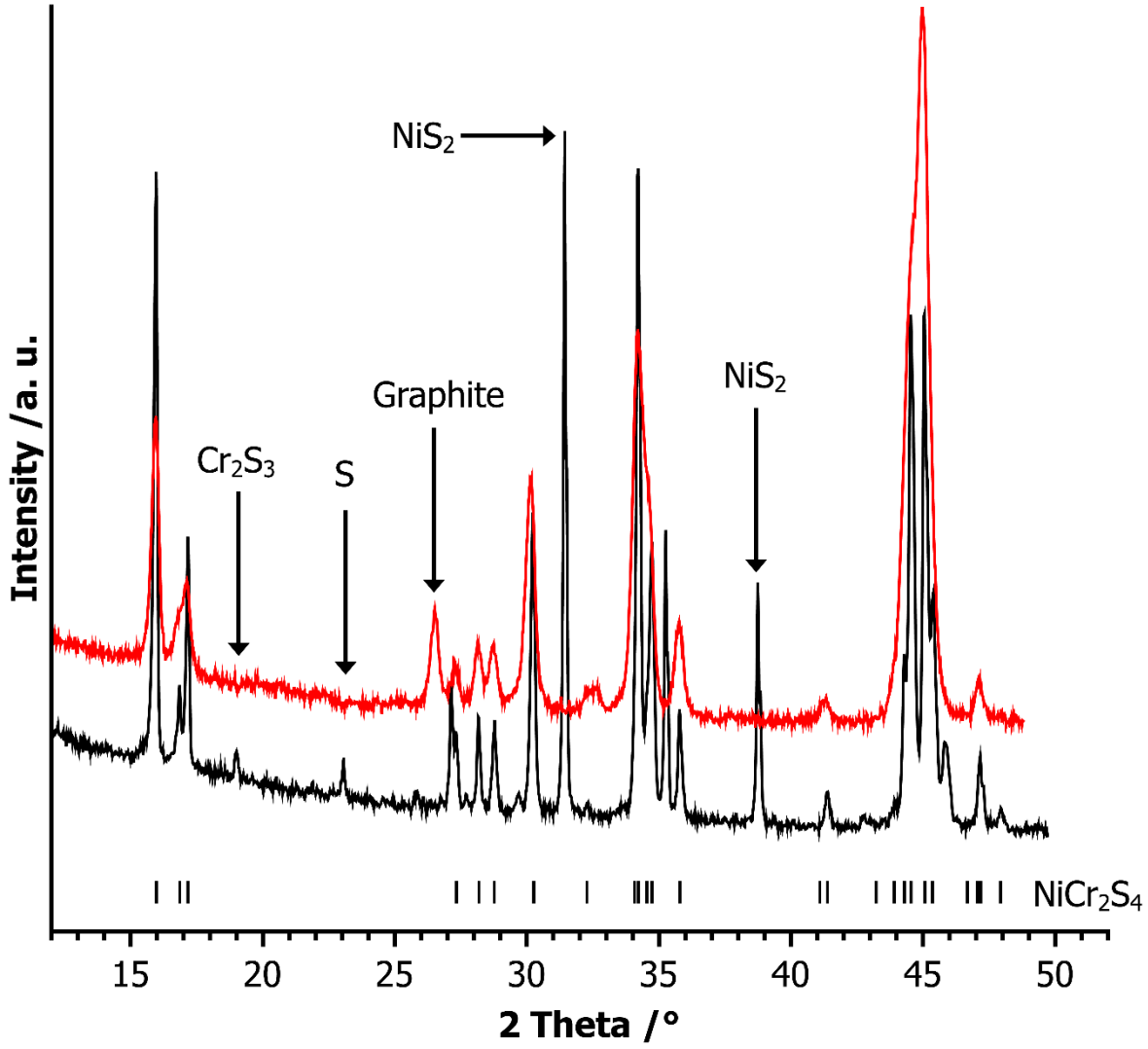

Figure S1: Room-temperature XRD pattern of $Ni_xCr_2S_4$ before (black) and after sintering (red). While the raw material prior to sintering contained traces of NiS_2, S and Cr_2S_3, these traces could be eliminated in the process and were no longer present in the post-sintering analysis. Additional reflections of C appeared, which were caused by residue from the sintering tools.

Table S1: EDX measurements of a large region and Cr and Ni rich domains. The regional scans were obtained from whole grains, while the domain compositions were obtained from point scans. Each composition given was averaged over four independent data points. Due to reasons of accuracy, the area scans were obtained with the EDX detector of a FIB-SEM system, while the domain scans were acquired in TEM.

Large area	**at%**
Ni	19(1)
Cr	22(1)
S	59(1)
Ni rich domain	**at%**
Ni	24(1)
Cr	20(1)
S	56(2)
Cr rich domain	**at%**
Ni	14(1)
Cr	27(1)
S	59(1)

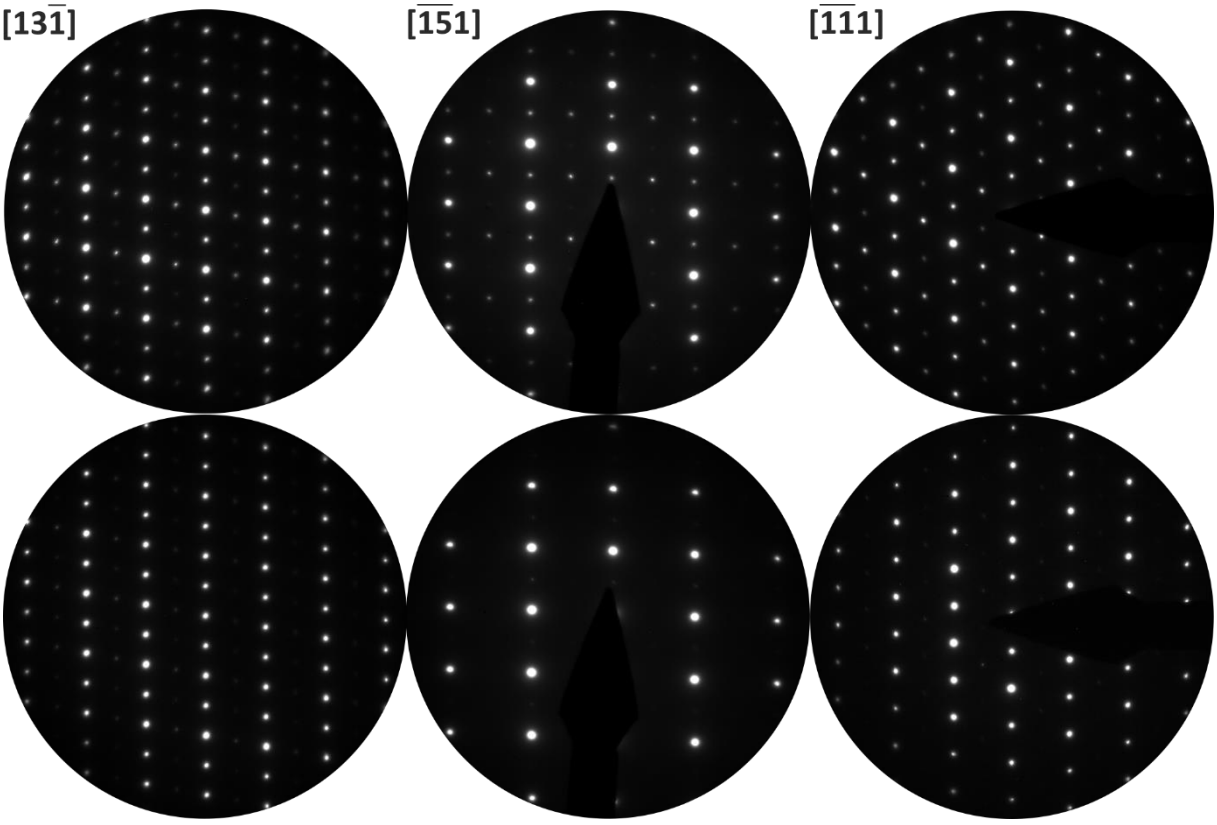

Figure S2: Experimental diffraction patterns of twinned $NiCr_2S_4$ structures in different zone axes obtained while keeping the sample cooled at 95 K. Diffraction patterns in the top row were acquired before, the bottom row after extensive electron beam irradiation. It is apparent that the superstructure reflections of the twin components vanished after irradiation, indicating that the cations reordered and caused the twin structures to disappear.

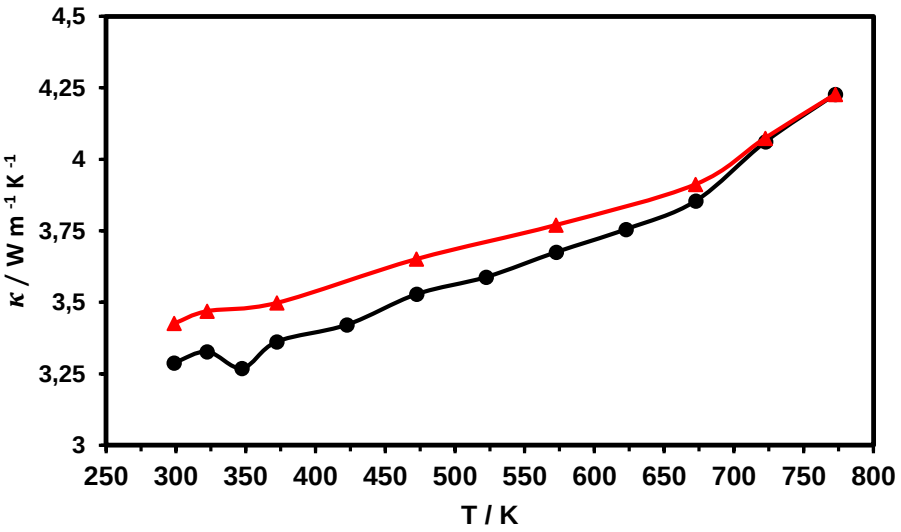

Figure S3: Temperature-dependent thermal conductivity measured while heating (black) and cooling (red). The lowest thermal conductivity at room temperature could be determined as 3.29 W m^{-1} K^{-1} while reaching its peak at 775 K and 4.23 W m^{-1} K^{-1}.