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# Show me your "Hand": Direct determination of "handedness" in $NaCu_5S_3$ chiral crystal via aberration-corrected scanning transmission electron microscopy

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Chirality (or handedness) is an important geometric characteristic of nature that is associated with a lack of center of inversion or any mirror symmetry and can be expressed as observables at various length scales. For example, in inorganic crystals, chirality can unlock particular properties, such as optical activity, piezoelectricity, chiral dichroism, enantioselective separations, and catalysis[1,2]. Determination of the handedness is of interest, in particular, at sub-micron particles and/or crystals containing defects. However, the determination of chirality for solid crystals is a challenging task, especially for crystals that have large anisotropy and have low dimension along one specific crystallographic direction. Here, we apply aberration-corrected scanning transmission electron microscopy (STEM) to determine the handedness of ferri-chiral (FEC) $NaCu_5S_3$ crystals by directly imaging the structure at an atomic scale. $NaCu_5S_3$ possesses the structural symmetries required to enable a special Dresselhaus spin splitting (D-1B) effect[3] with potential applications in spintronics and optoelectronics.

There exist two possible enantiomorphic forms for $NaCu_5S_3$, namely FEC-1 and FEC-2, as shown in Figure 1. The six-fold rotation of helices with different handedness show the relationship between the two configurations. Figure 1 shows the crystal structures of two configurations of $NaCu_5S_3$ along [210] and the one after rotation by 30° ([100] zone axis). The two configurations can be distinguished by comparing the atomic positions along those two zone axes. The atomic columns marked with the same oriented wedges in the [100] zone axis show different orientation along the [210] zone axis in two configurations: in FEC-1, sulfur atoms are on the right of copper atoms above the referred orange line, and sulfur atoms are on the left of copper atoms below the orange line; in FEC-2, sulfur atoms are on the left of copper atoms above the referred orange line, and sulfur atoms are on the right of copper atoms below the orange line.

In order to investigate the chirality of $NaCu_5S_3$ (space group *$P6_322$, #182)*, single crystals were synthesized by a facile solvothermal method[4,5] and its atomic structure was studied by high-resolution STEM (HRSTEM). Figure 2 displays a set of HRSTEM images of one crystal along the [210] zone axis and after rotation by 30°. Two images are aligned according to the edge shape of the particle to make sure the position of each atomic column is tracked after rotation. The white line marks the same atomic row before and after tilting. The inserted images show the corresponding high-magnified high-angle annular dark-field (HAADF) HRSTEM from the areas of interest. Comparing the orientation relationship of marked atoms, we can find it matches with the schematic of FEC-2 in Figure 1b, which reveals this particle is right-handed.

The presentation will cover similar examples of usage of aberration-corrected scanning transmission electron microscopy to determine the handedness of chiral crystals. We will also present complementary methods based on CBED to corroborate real-space/image analysis.[6]

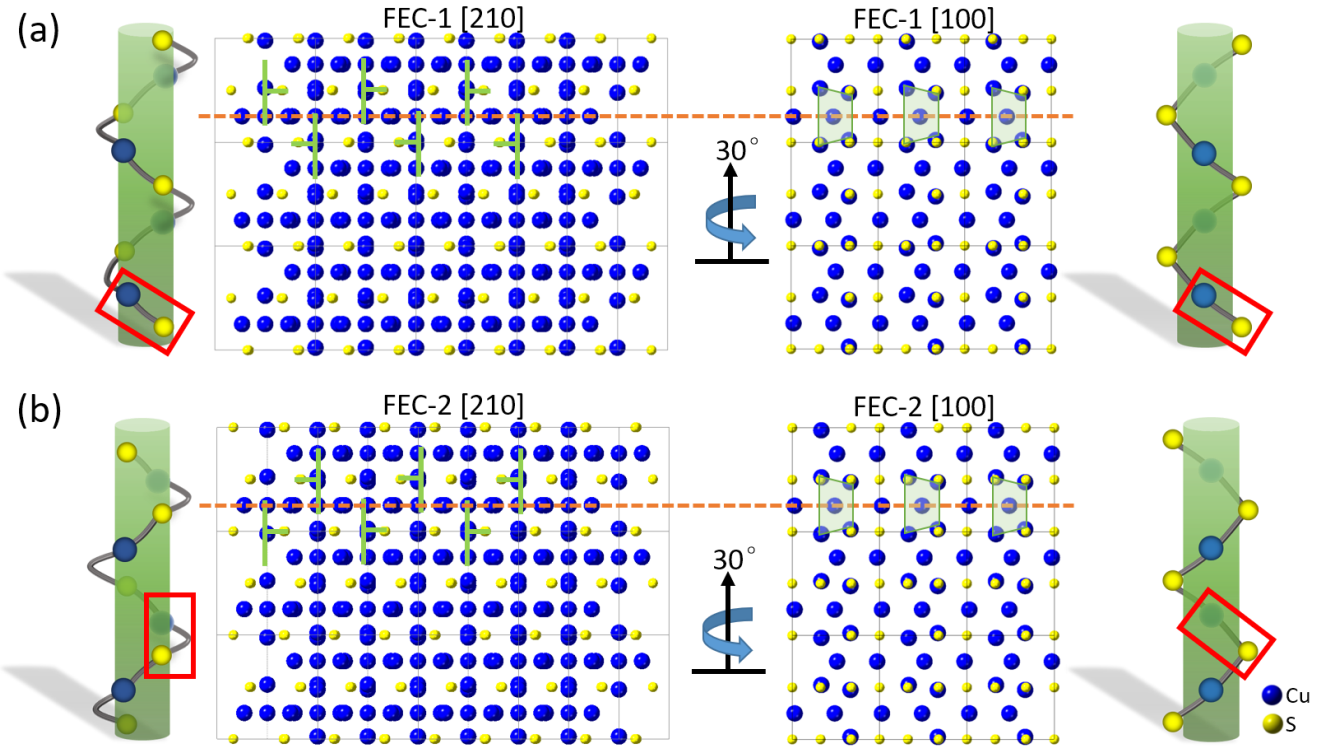


**Figure 1.** Crystal structure of two configurations of NaCu5S3, FEC-1 and FEC-2, viewed along [210] and [100] zone axes (displaying only copper – blue – and sulfur – yellow –atoms) together with the schematic representation of six-fold rotation of helices with different handedness. (a) Shows left-handed configuration (FEC-1) after rotation by 30°. (b) Shows right-handed configuration (FEC-2) after rotation by 30° along the same direction. The red rectangles circle two atomic columns which have the same orientation relationship in [100] zone axis, but different along [210] zone axis. The atomic columns marked with the same oriented wedges in the [100] zone axis show different orientation along [210] zone axis: in FEC-1, sulfur atoms are on the right of copper atoms above the referred orange line and sulfur atoms are on the left of copper atoms below the orange line; in FEC-2, sulfur atoms are on the left of copper atoms above the referred orange line, and sulfur atoms are on the right of copper atoms below the orange line.

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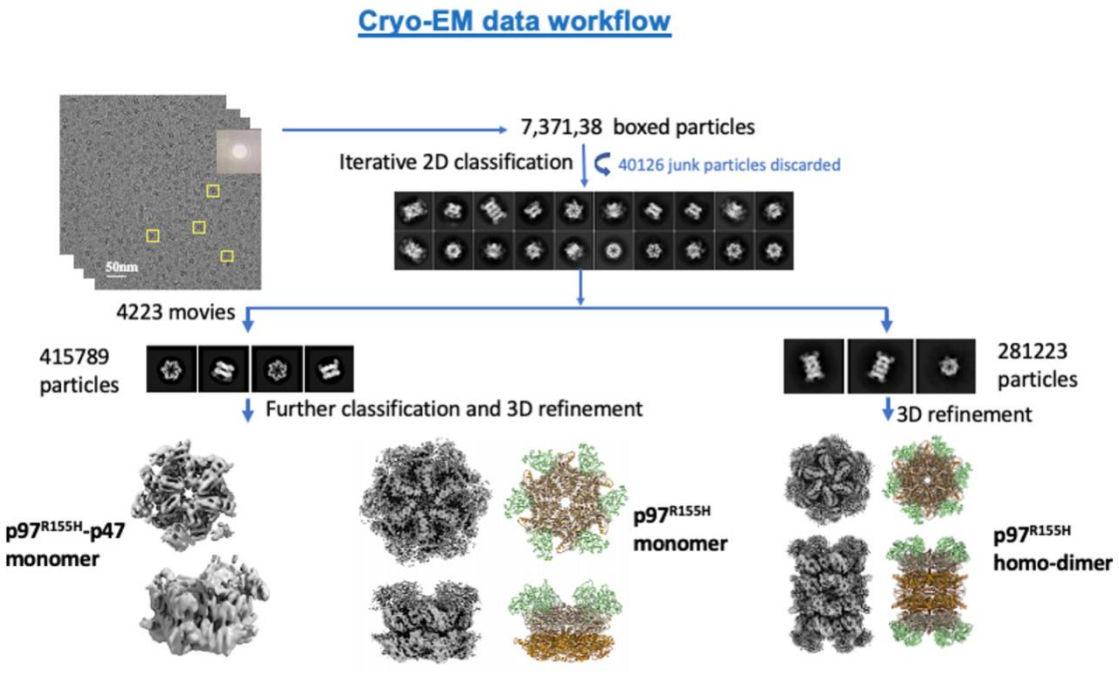


**Figure 2.** Determination of the handedness for the NaCu5S3 crystal from a set of HR-STEM images. Experimental micrographs and corresponding selected area electron diffraction (SAED) patterns taken along (a) [210] zone axis and (b) [100] zone axis. Two bright-field images are aligned according to the edge shape of the particle. The white line marks the same atomic row before and after tilting. The corresponding high-magnified HAADF-HRSTEM images inserted show the orientation relationship of marked atoms, which matches with the schematic of FEC-2 in Figure 1b, revealing a right-handed configuration.

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