On the Growth Morphology and Crystallography of Epitaxial CdTe/Cu$_7$Te$_4$ Interface

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Supplementary Materials

The detailed NCSL calculation process on the CdTe/Cu$_7$Te$_4$ interface

According to the definition of NCSL, the basal vectors of CdTe substrate $R_{S1}$, $R_{S2}$ and $R_{F1}$, $R_{F2}$ can be written as:

\[
R_{S1} = a_1 [1, 0, 0]_1, \quad R_{S2} = \frac{\sqrt{2}}{2} a_1 [0, 1, 0]_1 \\
R_{F1} = c [0, 1, 0]_1, \quad R_{F2} = \frac{\sqrt{3}}{2} a_2 [0, 0, 1]_1
\]

(S1)

(S2)

Here, $a_1$ and $a_2$, $c$ represents the lattice parameter of CdTe and Cu$_7$Te$_4$ respectively.

After the rotation process, the vectors $R_{RF1}$ and $R_{RF2}$ can also be obtained:
\[(R_{RF1}, R_{RF2}) = (Z/\theta)(R_{F1}, R_{F2}) \quad (S3)\]

Here, \((Z/\theta)\) represents the rotation matrix which can be expressed as:

\[
(Z/\theta) = \begin{bmatrix}
\cos \theta & -\sin \theta \\
\sin \theta & \cos \theta 
\end{bmatrix} \quad (S4)
\]

In this case, the Eq. (S3) can be written as:

\[
(R_{RF1}, R_{RF2}) = \begin{bmatrix}
\cos \theta & -\sin \theta \\
\sin \theta & \cos \theta 
\end{bmatrix} \begin{bmatrix}
c & 0 \\
0 & \sqrt{3} \over 2 a_2 
\end{bmatrix} = \begin{bmatrix}
c \cos \theta & -\sqrt{3} a_2 \sin \theta \\
c \sin \theta & \sqrt{3} a_2 \cos \theta 
\end{bmatrix} \quad (S5)
\]

Therefore,

\[
R_{RF1} = c \begin{bmatrix}
\cos \theta \\
\sin \theta 
\end{bmatrix}, \quad R_{RF2} = \sqrt{3} \over 2 a_2 \begin{bmatrix}
-\sin \theta \\
\cos \theta 
\end{bmatrix} \quad (S6)
\]

In this case, the NCSL basal vectors \(V_1\) and \(V_2\) can be expressed as:

\[
V_1 = mR_{S1} + nR_{S2} = a_1 \begin{bmatrix}
m \\
n 
\end{bmatrix} + \sqrt{3} \over 2 a_2 \begin{bmatrix}
m \over 2 \\
n \over 2 
\end{bmatrix} = a_1 \begin{bmatrix}
m \over 2 \\
n \over 2 
\end{bmatrix} \quad (S7)
\]

\[
V_2 = pR_{RF1} + qR_{RF2} = pc \begin{bmatrix}
\cos \theta \\
\sin \theta 
\end{bmatrix} + \sqrt{3} \over 2 qa_2 \begin{bmatrix}
-\sin \theta \\
\cos \theta 
\end{bmatrix} = \begin{bmatrix}
pc \cos \theta - \sqrt{3} qa_2 \sin \theta \\
pc \sin \theta + \sqrt{3} qa_2 \cos \theta 
\end{bmatrix} \quad (S8)
\]

Here, \(m, n, p\) and \(q\) are all integers. According to the definition of NCSL, \(V_1\) and \(V_2\) should be equal, the 2 vectors must have the same vector module and parallel to each other.

Therefore, combine the Eq. (S7) and (S8), the following equation can be obtained:

\[
\begin{cases}
ma_1 = pc \cos \theta - \sqrt{3} qa_2 \sin \theta \\
\sqrt{2} \over 2 ma_1 = pc \sin \theta + \sqrt{3} qa_2 \cos \theta
\end{cases} \quad (S9)
\]
From Eq. (S9), the expressions of $\tan \theta$, $\Sigma$ and the relationship between the 4 integers can be obtained accordingly:

\[
\begin{align*}
    &\begin{aligned}
        m^2a_1^2 + \frac{1}{2}n^2a_1^2 = p^2c^2 + \frac{3}{4}q^2a_2^2 \\
        \tan \theta = \frac{2\sqrt{2}npc - 2\sqrt{3}mqa_2}{4mpc + \sqrt{6}nqa_2} \\
        \Sigma = \frac{V_{\text{Super}}}{V_{\text{Unit}}} = \frac{4p^2c^2 + 3q^2a_2^2}{2\sqrt{3}a_2c}
    \end{aligned}
\end{align*}
\]

(S10)

According to the TEM results in Fig. 2, the parallel relationship of $\{1\overline{1}1\}_{CdTe}/\{0001\}_{Cu_7Te_4}$ can be obtained. In addition, it is also noticed that the interplanar distance of $\{0001\}_{Cu_7Te_4}$ (0.7211 nm) is very close to that of $\{0\overline{1}10\}_{Cu_7Te_4}$ (0.7218 nm). This means that the established Cu$_7$Te$_4$ unit cell is similar to square. To simplify the calculation, the following relations can be set:

\[
\begin{align*}
    \sqrt{3}a_1 &= \frac{1}{2}c \\
    c &= \sqrt{3} \frac{a_2}{2}
\end{align*}
\]

(S11)

In this case, Eq. (S10) can be simplified to be:

\[
\begin{align*}
    \begin{aligned}
        &\frac{3}{4}m^2 + \frac{3}{8}n^2 = p^2 + q^2 \\
        \tan \theta = \frac{\sqrt{2}np - 2mq}{2mp + \sqrt{2}nq} \\
        \Sigma = \frac{V_{\text{Super}}}{V_{\text{Unit}}} = p^2 + q^2
    \end{aligned}
\end{align*}
\]

(S12)

Under the principle that the value of the 4 integers should be as small as possible, the calculation results are summarized in Table S1. The rotation angle of $\pm54.74^\circ$ and $\pm35.26^\circ$ refer to the variant 1 and 2 of Cu$_7$Te$_4$ respectively. The same $\Sigma$ values suggest the equivalent status of the COR obtained by applying the rotation along the 4 rotation angles.
Table S1 The calculation results of the CdTe/Cu$_7$Te$_4$ interface

<table>
<thead>
<tr>
<th>$m$</th>
<th>$n$</th>
<th>$p$</th>
<th>$q$</th>
<th>tan$\theta$</th>
<th>$\theta$</th>
<th>$\Sigma$</th>
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<tr>
<td>2</td>
<td>4</td>
<td>3</td>
<td>0</td>
<td>$\frac{\sqrt{2}}{2}$</td>
<td>54.74°</td>
<td>9</td>
</tr>
<tr>
<td>-2</td>
<td>4</td>
<td>3</td>
<td>0</td>
<td>$-\frac{\sqrt{2}}{2}$</td>
<td>-54.74°</td>
<td>9</td>
</tr>
<tr>
<td>2</td>
<td>4</td>
<td>0</td>
<td>3</td>
<td>$\frac{\sqrt{2}}{2}$</td>
<td>-35.26°</td>
<td>9</td>
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<td>-2</td>
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