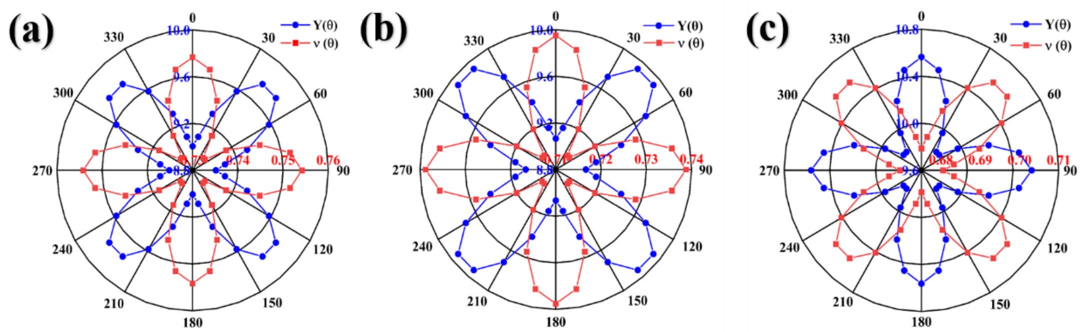
Supplementary Information (SI) for Physical Chemistry Chemical Physics.

Supporting Information

Furthermore, the Young's modulus and Poisson's ratio are calculated using the elastic constant according to the formula:

$$\begin{cases} Y(\theta) = \dfrac{C_{11}C_{22} - C_{12}^2}{C_{11}sin^4\theta + C_{22}cos^4\theta + \left(\dfrac{C_{11}C_{22} - C_{12}^2}{C_{66}} - 2C_{12}\right)sin^2\theta cos^2\theta} \\ v(\theta) = -\dfrac{\left(C_{11} + C_{22} - \dfrac{C_{11}C_{22} - C_{12}^2}{C_{66}}\right)sin^2\theta cos^2\theta - C_{12}(sin^4\theta + cos^4\theta)}{C_{11}sin^4\theta + C_{22}cos^4\theta + \left(\dfrac{C_{11}C_{22} - C_{12}^2}{C_{66}} - 2C_{12}\right)sin^2\theta cos^2\theta} \end{cases} \quad (1)$$

The blue and red lines represent the changes in Young's modulus and Poisson's ratio along the inplane 0-360° direction, respectively, as shown in Figure 2(a)-(c). It can be clearly seen that SL-Tl*X* (*X*=Cl/Br/I) has anisotropic Young's modulus and Poisson's ratio. It is worth noting that the Young's modulus of SL-TlCl and SL-TlBr reach their minimum values and maximum values in the 0° and 45° directions, respectively, while the TlI has the opposite trend, with the Young's modulus reaching their



minimum values and maximum values in the 45° and 0° directions, respectively. The

Figure S1. (a)-(c) The radar plot of Young's modulus and Poisson's ratio of SL-TlCl, SL-TlBr and SL-TlI. The blue and red lines represent the Young's modulus and Poisson's ratio, respectively.

change trend of Poisson's ratio is opposite to that of Young's modulus for all the three materials. SL-TlCl and SL-TlBr reach their minimum values and maximum values in the 45° and 0° directions, respectively, and SL-TlI reach their minimum values and maximum values in the 0° and 45° directions, respectively. For SL-TlCl and SL-TlBr, the Young's modulus reaching their minimum values in the 0°direction, indicating that the two SLs are more prone to deformation along the *x* or *y* direction. However, for the two directions, the stress required for uniaxial strain to occur in SL-TlI is greater than SL-TlCl and SL-TlBr.

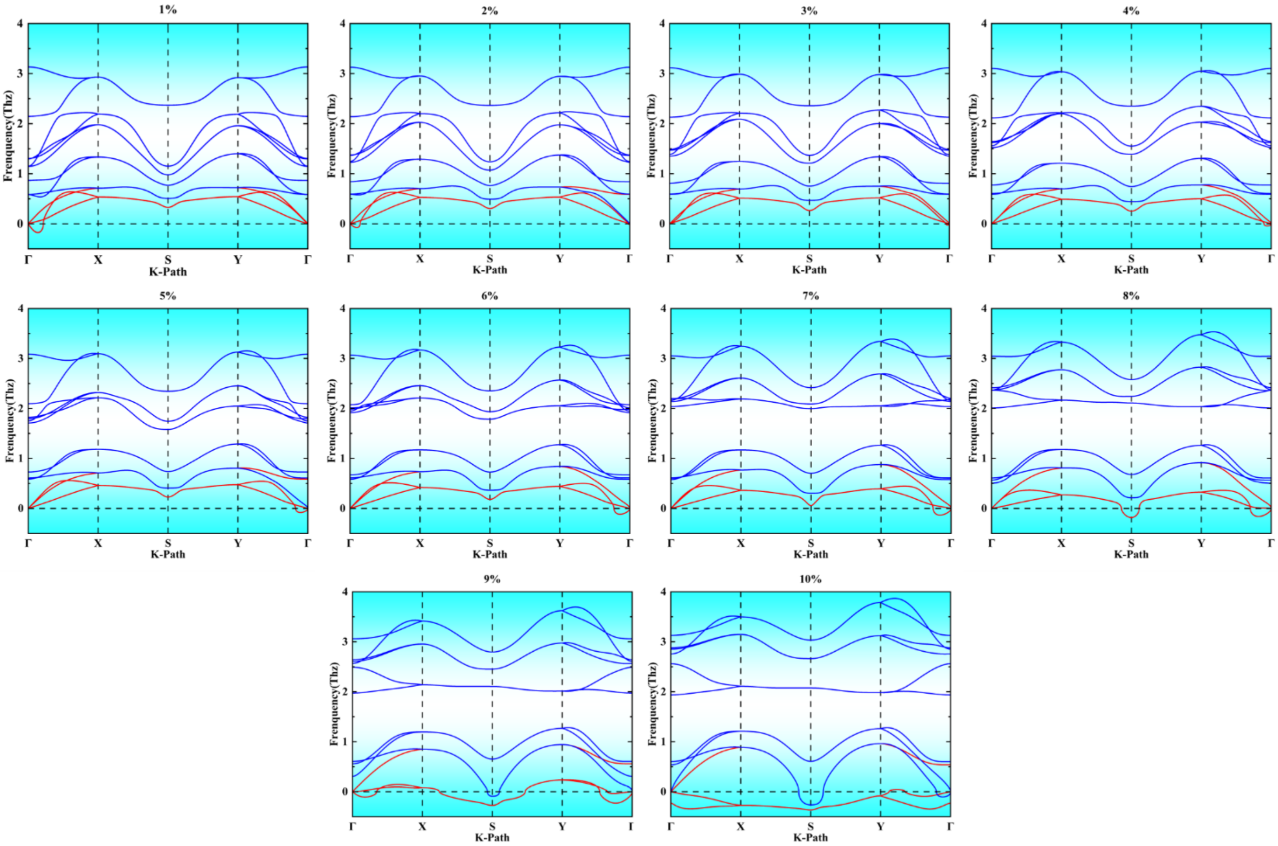


Figure S2. The phonon spectrum of TlCl with the atomic relative displacement of 1%~10%.

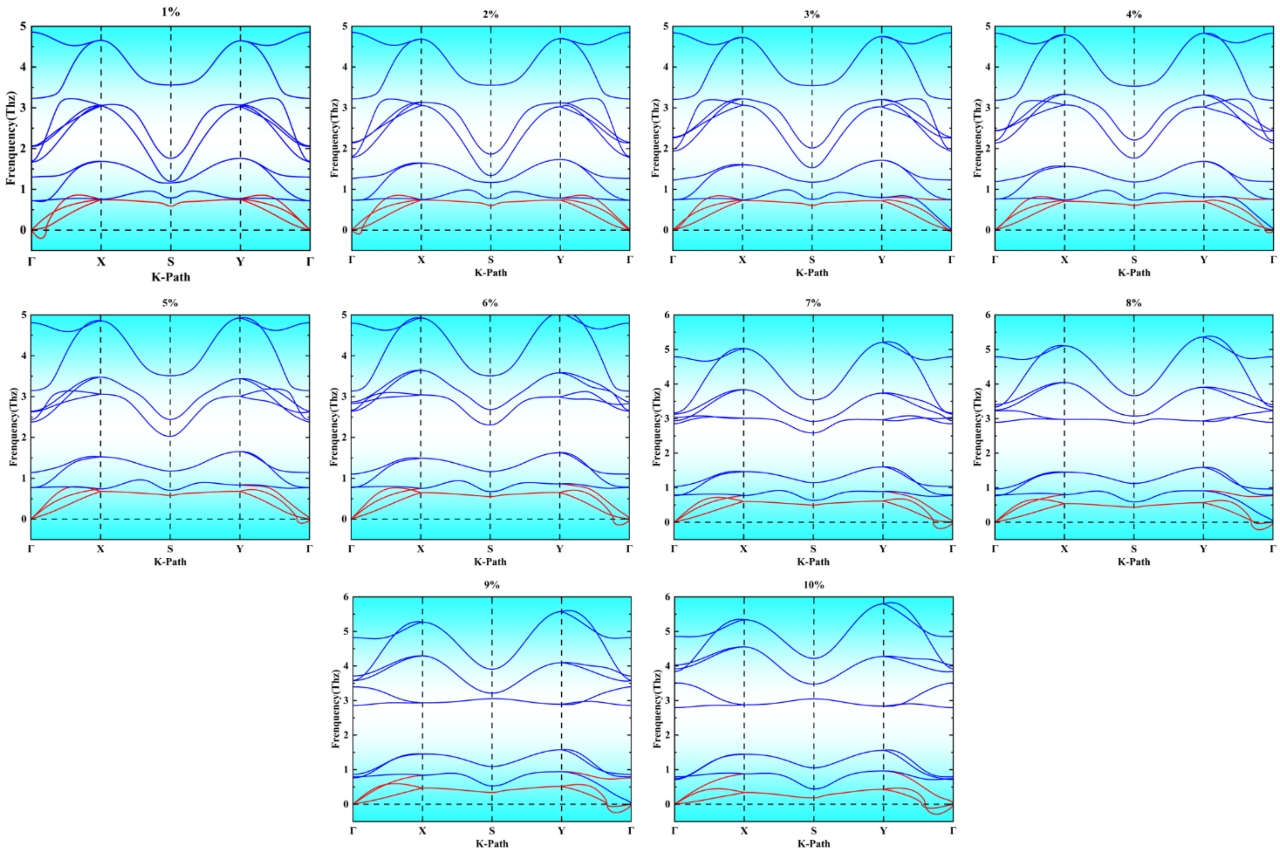


Figure S3. The phonon spectrum of TlBr with the atomic relative displacement of 1%~10%.

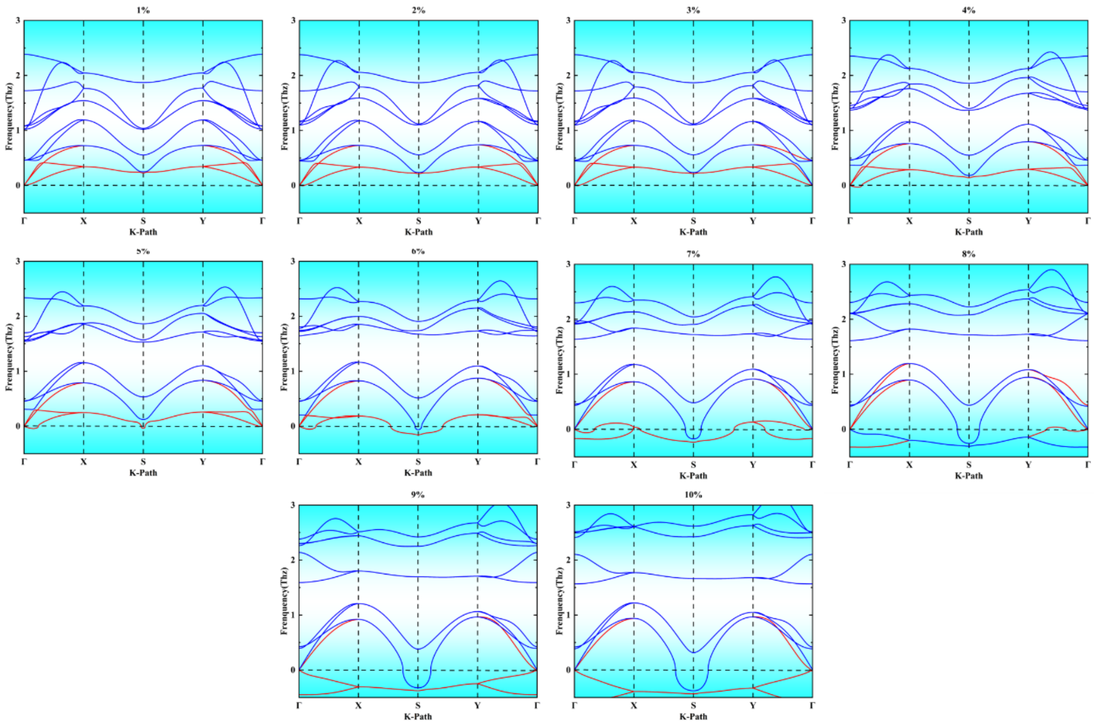


Figure S4. The phonon spectrum of TlI with the atomic relative displacement of 1%~10%.

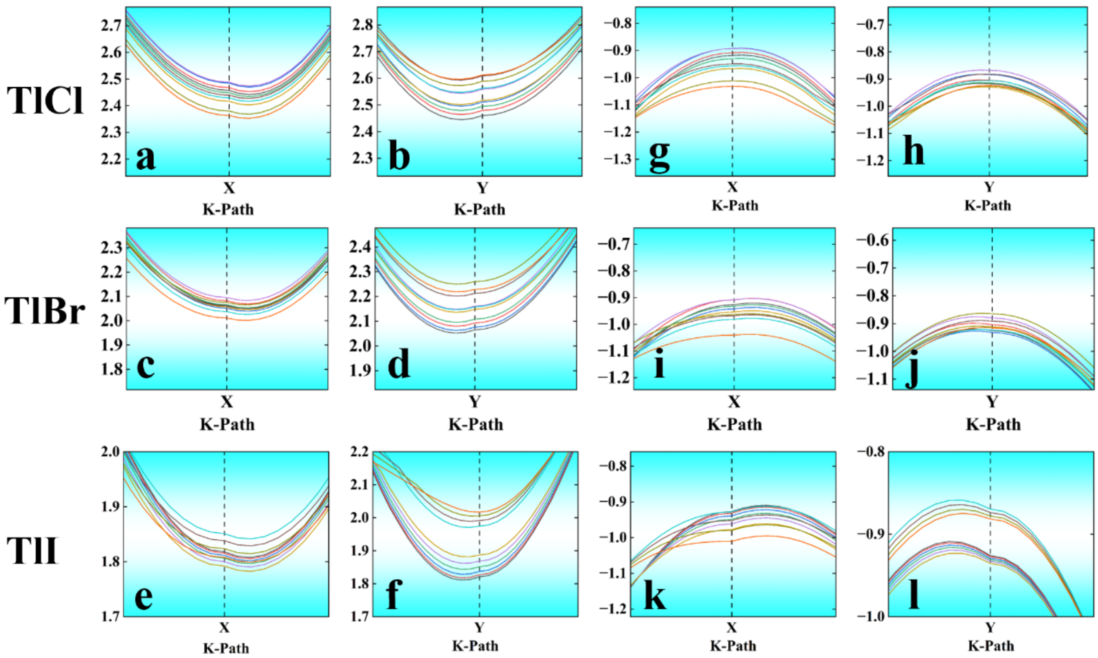


Figure S5. (a)-(h) The drawing of partial enlargement of the conduction band and the valence band on the X- and Y- points of SL-TlCl and SL-TlBr with relative displacement of 1%~10%.

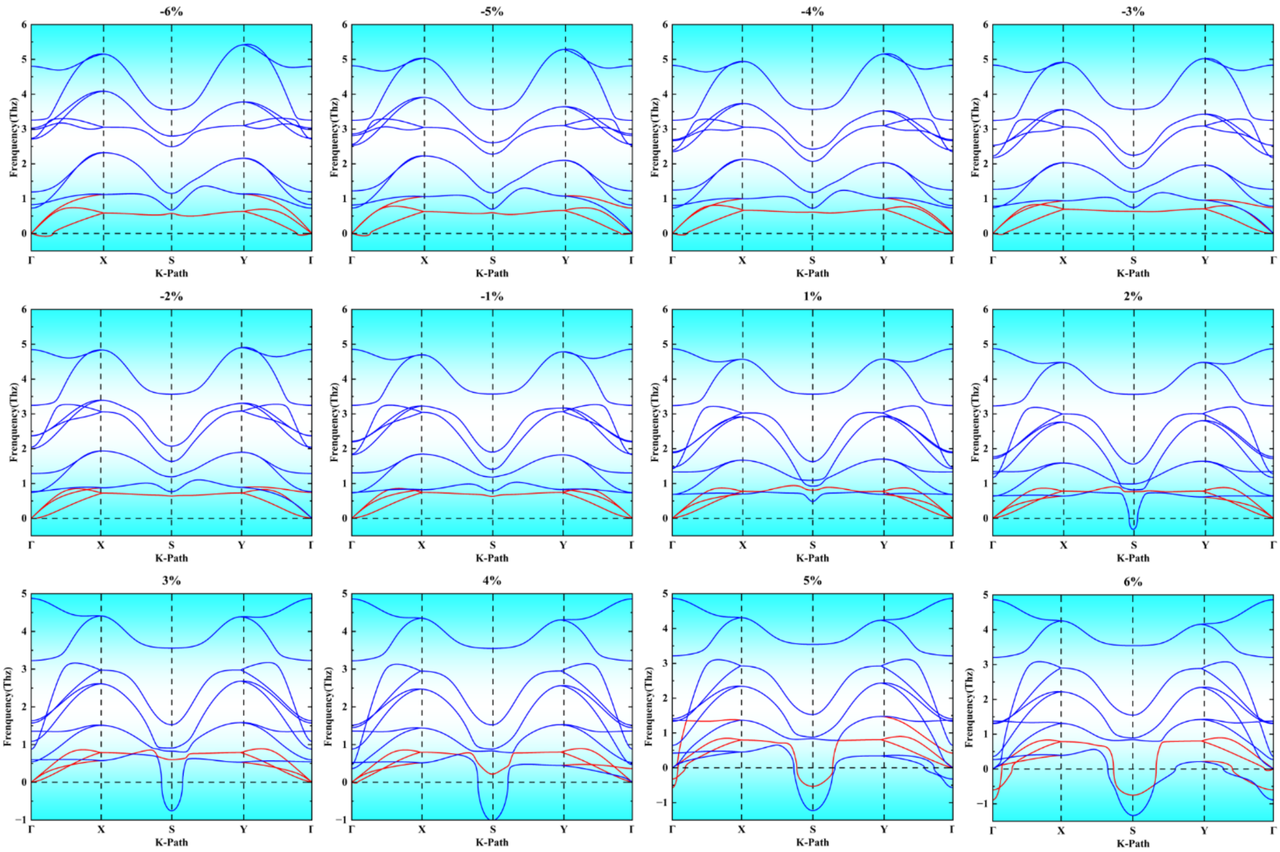


Figure S6. The phonon spectrum of TlCl with the uniaxial strain of -6%~6%.

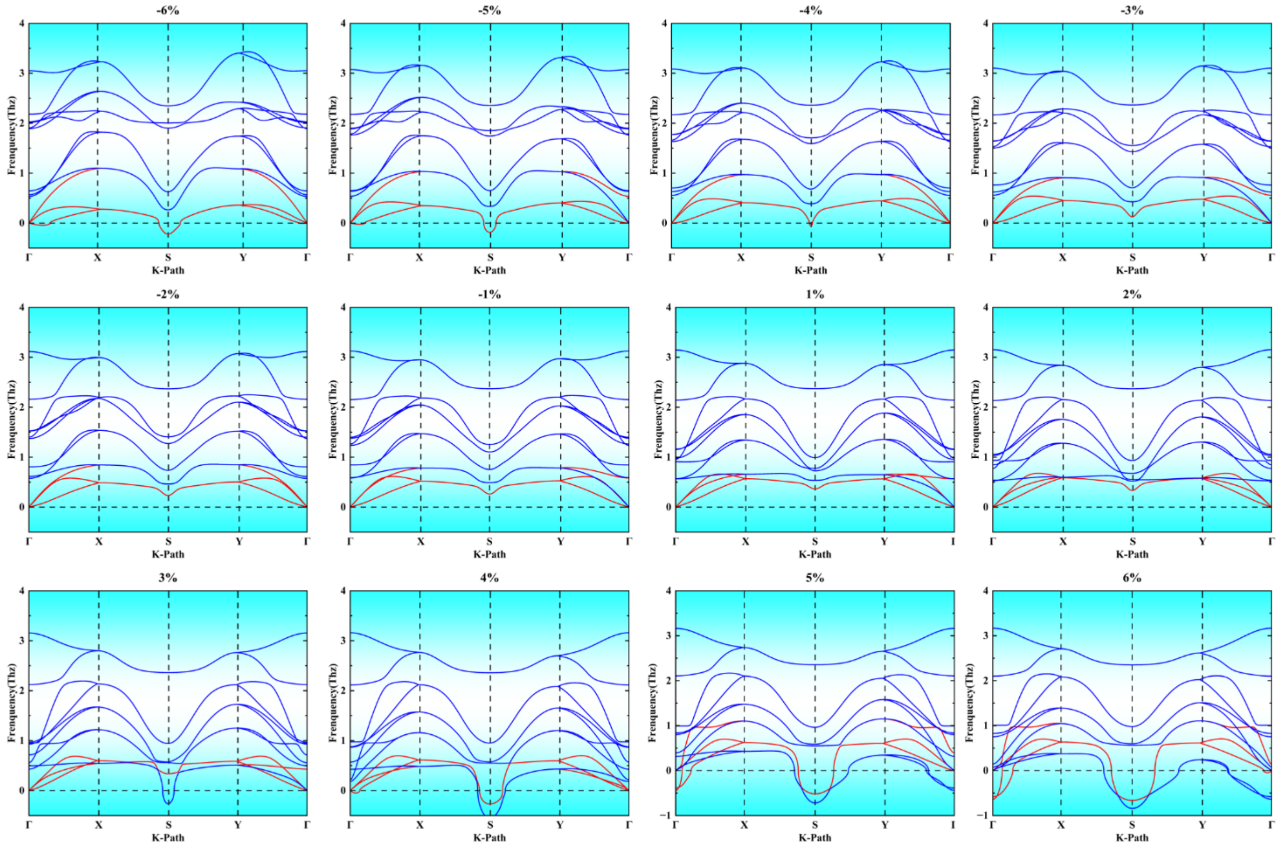


Figure S7. The phonon spectrum of TlBr with the uniaxial strain of -6%~6%.

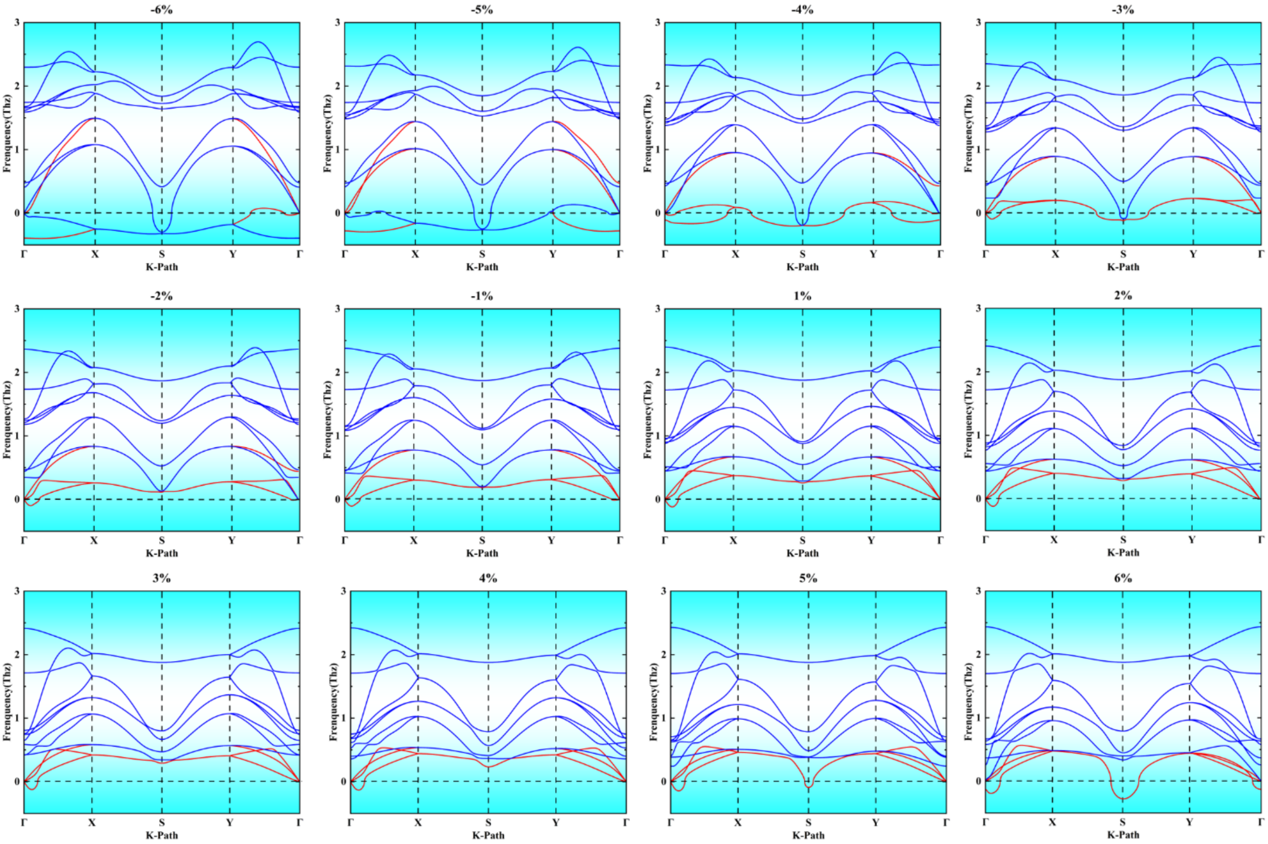


Figure S8. The phonon spectrum of TlI with the uniaxial strain of -6%~6%.