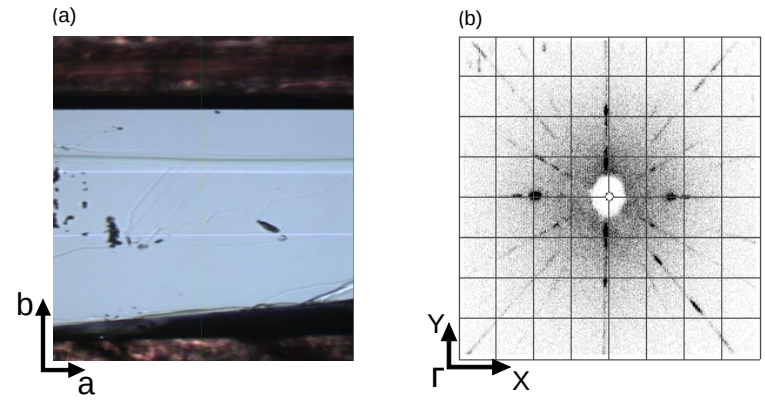
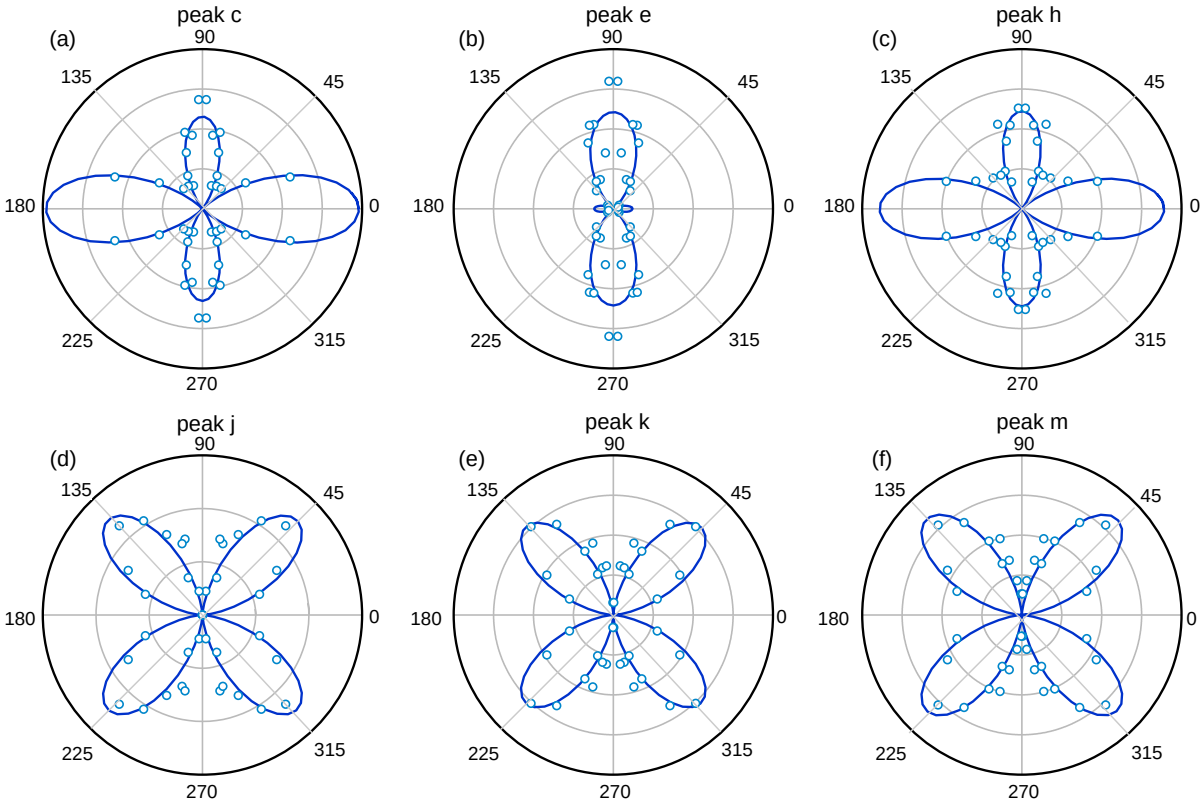
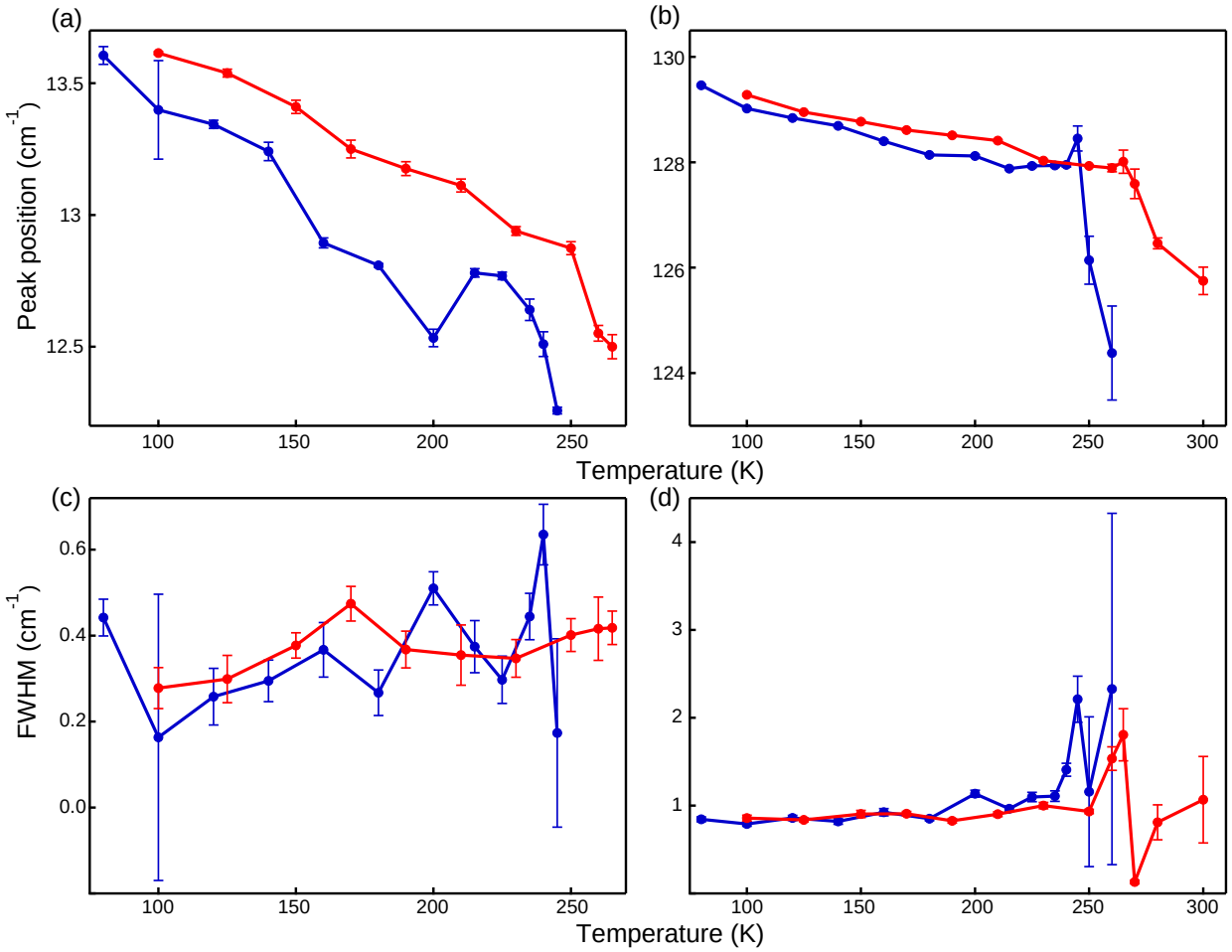


Supplementary Figure 1: Photograph and Laue pattern of the sample. (**a**) Photograph of the sample. (**b**) The Laue pattern of the sample in the ab plane.



Supplementary Figure 2: Angular dependence of the Raman intensities for three A_g modes c, e, h, and three B_g modes j, k, m measured at HH configuration. The solid curves are fits using the corresponding cross sections above. The numbers on the angular axis indicate the light polarization angle relative to the a-axis of crystal, as denoted by θ.



Supplementary Figure 3: Peak position and FWHM of peak A (a) (c) and peak D (b) (d) extracted from the fits versus temperature. The blue and red lines denote cooling down and warming up respectively. The error bar is defined by the uncertainty from the Lorentz fitting.

Supplementary Table 1: Raman selection rules in $1T'$ phase $MoTe_2$.

	-x(yy)x	-x(yz)x	-x(zz)x	-y(xx)y	-y(xz)y	-y(zz)y	-z(xx)z	-z(xy)z	-z(yy)z
A_g	Y	N	Y	Y	N	Y	Y	Y	Y
B_g	N	Y	N	N	Y	N	N	N	N

Supplementary Table 2: Raman selection rules in T_d phase $MoTe_2$.

	-x(yy)x	-x(yz)x	-x(zz)x	-y(xx)y	-y(xz)y	-y(zz)y	-z(xx)z	-z(xy)z	-z(yy)z
A_1	Y	N	Y	Y	N	Y	Y	N	Y
A_2	N	N	N	N	N	N	N	Y	N
B_1	N	N	N	N	Y	N	N	N	N
B_2	N	Y	N	N	N	N	N	N	N

Supplementary Note 1: The photo and Laue pattern of the sample

Single crystal samples were oriented by Laue diffraction patterns. Supplementary Figure 1(a) shows a photo of our sample. Supplementary Figure 1(b) shows the corresponding Laue pattern of the sample in ab plane.

Supplementary Note 2: The Raman selection rules in 1T' and T_d phase $MoTe_2$

In this article, we define the direction of the molybdenum atom chains as a-axis for both high temperature 1T' $MoTe_2$ phase and low temperature T_d phase. So we describe the monoclinic 1T' phase in the space group $P2_1/m11$. From this arrangement, we can determine that the screw axis is a, and the mirror plane is bc. So we define that x=b, y=c, z=a, and the Raman selection rules are listed in the Supplementary Table 1 (The letters outside of parenthesis denote the propagation directions of incident and reflected light respectively. The letters inside of parenthesis denote polarization directions of incident and reflected light. Y/N denote the mode can/cannot be observed by Raman spectrum in this configurations.).

T_d phase $MoTe_2$ belongs to space groups $Pmn2_1$. The screw axis is c, glide plane is ac and mirror plane is bc. We define that x=a, y=b, z=c, and the selection rules are listed in the Supplementary Table 2.

Supplementary Note 3: Angular dependence of the Raman intensities

In the 1T′ structure, Raman tensor for the A_g and B_g modes of C_{2h} point group are given by

$$R(A_g) = \begin{pmatrix} a & d & 0 \\ d & b & 0 \\ 0 & 0 & c \end{pmatrix} \qquad R(B_g) = \begin{pmatrix} 0 & 0 & e \\ 0 & 0 & f \\ e & f & 0 \end{pmatrix} \tag{1}$$

The Raman cross-section is $I = | < p_i|R|p_r > |^2$, where p_i and p_r are polarization states of the incident and scattered light respectively. For the HH configuration on ab plane, θ is the angle between a-axis and the polarization of incident light. The Raman cross sections for A_g and B_g can be calculated:

$$I(A_g) = (a \cdot cos^2(\theta) + c \cdot sin^2(\theta))^2 \tag{2}$$

$$I(B_g) = e^2 \cdot sin^2(2\theta) \tag{3}$$

In HH configuration, A_g mode shows two-fold symmetry while B_g mode shows four-fold symmetry. The excellent agreement between data and theoretical calculation verifies the symmetry properties of the A_g, B_g modes, and confirms the accuracy of polarizations.

Supplementary Note 4: Full width at half maximum (FWHM) and Raman frequency of Peak A and D

The symmetric Lorentzian lineshapes at different temperatures suggest that there is phonon-phonon coupling in these phonon modes probed in our experiments. We find that the FWHM of peak A and D does not change substantially, which suggests that phonon-phonon interactions are weak for these two modes.

The peak position of these two Raman peaks soften with temperature increasing, and can be described by the linear Gruneisen model $\omega(T) = \omega_0 + \chi T$. $\omega(T)$ is the temperature-dependent phonon frequency, ω_0 is the harmonic phonon frequency at 0 K, and χ is the first order temperature coefficient[1]. The deviation of peak position of these two modes from linear temperature dependence at T $\approx$ 260 K upon warming and $\approx$ 250 K upon cooling, indicates the phase transition.

Supplementary Reference

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[1] Jana, M. K. *et al.* A combined experimental and theoretical study of the structural, electronic and vibrational properties of bulk and few-layer T_d-WTe_2. *Journal of Physics Condensed Matter* **27**, 39–67 (2015).