Supplementary Information

Significantly anisotropic Raman response of (110) surface in quasi-one-dimensional system $(TaSe_4)_2I$ by polarized Raman spectroscopy

Xiao Gao[a], Pin Song[b], Zhongti Sun[c], Haiping Chen[a]

[a]*School of Mechanics and Optoelectronic Physics, Anhui University of Science and Technology, Huainan 232001, P. R. China. E-mail: hpchen01@mail.ustc.edu.cn*

[b]*Key Laboratory of Functional Molecular Solids, Ministry of Education, College of Chemistry and Materials Science, Anhui Normal University, Wuhu 241000, P. R. China. songpin@ahnu.edu.cn*

[c]*School of Materials Science and Engineering, Jiangsu University, Zhenjiang, Jiangsu, 212013, P. R. China. ztsun@ujs.edu.cn*

To whom correspondence should be addressed. E-mail: hpchen01@mail.ustc.edu.cn

The transformation matrix from [001]-axis to [110]-axis as follows

$$\begin{pmatrix} \frac{1}{2} & -\frac{1}{2} & \frac{\sqrt{2}}{2} \\ -\frac{1}{2} & \frac{1}{2} & \frac{\sqrt{2}}{2} \\ -\frac{\sqrt{2}}{2} & -\frac{\sqrt{2}}{2} & 0 \end{pmatrix}$$

The inverse transformation matrix from [001]-axis to [110]-axis as follows

$$\begin{pmatrix} \frac{1}{2} & -\frac{1}{2} & -\frac{\sqrt{2}}{2} \\ -\frac{1}{2} & \frac{1}{2} & -\frac{\sqrt{2}}{2} \\ \frac{\sqrt{2}}{2} & \frac{\sqrt{2}}{2} & 0 \end{pmatrix}$$

When the exposed crystal surface is (110), The Raman tensors are given below:

$$\begin{pmatrix} \frac{1}{2} & -\frac{1}{2} & -\frac{\sqrt{2}}{2} \\ -\frac{1}{2} & \frac{1}{2} & -\frac{\sqrt{2}}{2} \\ \frac{\sqrt{2}}{2} & \frac{\sqrt{2}}{2} & 0 \end{pmatrix} \begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \begin{pmatrix} \frac{1}{2} & -\frac{1}{2} & \frac{\sqrt{2}}{2} \\ -\frac{1}{2} & \frac{1}{2} & \frac{\sqrt{2}}{2} \\ -\frac{\sqrt{2}}{2} & -\frac{\sqrt{2}}{2} & 0 \end{pmatrix}$$

Where the intermediate 3×3 matrixes are the Raman tensors， which corresponds to

C_4 rotating axis paralleling with z-axis:

$A_1 \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix}$ $B_1 \begin{pmatrix} c & 0 & 0 \\ 0 & -c & 0 \\ 0 & 0 & 0 \end{pmatrix}$ $B_2 \begin{pmatrix} 0 & d & 0 \\ d & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$ $E(-y) \begin{pmatrix} 0 & 0 & e \\ 0 & 0 & 0 \\ e & 0 & 0 \end{pmatrix}$ $E(x) \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & e \\ 0 & e & 0 \end{pmatrix}$

So the obtained Raman tensors of exposed (110) crystal surface are given below:

$$A_1 \begin{pmatrix} \frac{1}{2}(a+b) & -\frac{1}{2}(a-b) & 0 \\ -\frac{1}{2}(a-b) & \frac{1}{2}(a+b) & 0 \\ 0 & 0 & a \end{pmatrix}$$

$$B_1 \begin{pmatrix} 0 & 0 & \frac{\sqrt{2}}{2}c \\ 0 & 0 & -\frac{\sqrt{2}}{2}c \\ \frac{\sqrt{2}}{2}c & -\frac{\sqrt{2}}{2}c & 0 \end{pmatrix}$$

$$B_2 \begin{pmatrix} -\frac{1}{2}d & \frac{1}{2}d & 0 \\ \frac{1}{2}d & -\frac{1}{2}d & 0 \\ 0 & 0 & d \end{pmatrix}$$

$$E(-y) \begin{pmatrix} -\frac{\sqrt{2}}{2}e & 0 & -\frac{1}{2}e \\ 0 & \frac{\sqrt{2}}{2}e & -\frac{1}{2}e \\ -\frac{1}{2}e & -\frac{1}{2}e & 0 \end{pmatrix}$$

$$E(x) \begin{pmatrix} \frac{\sqrt{2}}{2}e & 0 & -\frac{1}{2}e \\ 0 & -\frac{\sqrt{2}}{2}e & -\frac{1}{2}e \\ -\frac{1}{2}e & -\frac{1}{2}e & 0 \end{pmatrix}$$

The density functional perturbation theory calculation gives the following Raman tensors

Mode A_1

Frequency(THZ) 2.95521506

Raman tensor:

$$\begin{pmatrix} 20481.82806681 & -19586.23583943 & 0.2346199 \\ 19586.22960369 & -22359.51139662 & -0.2738533 \\ 0.23877706 & -0.26943614 & -1811.94766639 \end{pmatrix}$$

Mode A_1

Frequency(THZ) 4.30162995

Raman tensor:

$$\begin{pmatrix} 80250.68156099 & 76629.33045460 & -0.26241946 \\ 76629.36575534 & 87595.62690999 & -0.50381421 \\ -0.23802042 & -0.47837691 & 7205.84979990 \end{pmatrix}$$

Mode A_1

Frequency(THZ) 4.76882955

Raman tensor:

$$\begin{pmatrix} -79686.18914718 & -76538.70749079 & 13.36109830 \\ -76538.76101313 & -87022.74886801 & -12.17737125 \\ 13.32420426 & -12.21582419 & 6727.87536895 \end{pmatrix}$$

Mode A_1

Frequency(THZ) 5.45630113

Raman tensor:

$$\begin{pmatrix} 17756.42617598 & 18602.77347959 & 11.46494658 \\ 18602.82855956 & 19540.22746136 & -10.64810032 \\ 11.50313882 & -10.60808940 & 24.15724106 \end{pmatrix}$$

Mode A_1

Frequency(THZ) 5.57229905

Raman tensor:

$$\begin{pmatrix} -14365.82238055 & -16155.30792731 & -10.79310916 \\ -16155.30766749 & -15913.64439316 & 10.90223544 \\ -10.79310916 & 10.90223544 & 1034.10015624 \end{pmatrix}$$

Mode A_1

Frequency(THZ) 8.42015967

Raman tensor:

$$\begin{pmatrix} 27053.20840431 & 28621.99089474 & 0.01740794 \\ 28622.00544466 & 29796.85177861 & -0.14835719 \\ 0.02754092 & -0.13770458 & -229.82374045 \end{pmatrix}$$

Mode B_1

Frequency(THZ) 1.93192414

Raman tensor:

$$\begin{pmatrix} -11.15436508 & -11.43336976 & 45998.59567938 \\ -11.47808504 & -14.50964094 & -43848.15938841 \\ 45998.56493763 & -43848.19199330 & -1.3090862 \end{pmatrix}$$

Mode B_1

Frequency(THZ) 2.21425513

Raman tensor:

$$\begin{pmatrix} -11.15436508 & -11.43336976 & 45998.59567938 \\ -11.47808504 & -14.50964094 & -43848.15938841 \\ 45998.56493763 & -43848.19199330 & -1.3090862 \end{pmatrix}$$

Mode B_1

Frequency(THZ) 4.17334741

Raman tensor:

$$\begin{pmatrix} 4.11104601 & 2.94094935 & -29420.11684859 \\ 2.95871711 & 2.40868928 & 28042.36666643 \\ -29420.10448841 & 28042.37979912 & 0.35406754 \end{pmatrix}$$

Mode B_1

Frequency(THZ) 4.72524990

Raman tensor:

$$\begin{pmatrix} -1.98032297 & -0.26557740 & 3370.48771958 \\ -0.23428736 & 1.80628832 & -3210.19818525 \\ 3370.50944158 & -3210.17542886 & 0.07214809 \end{pmatrix}$$

Mode B_1

Frequency(THZ) 5.07146425

Raman tensor:

$$\begin{pmatrix} -10.12542546 & -6.52335788 & 37260.03971529 \\ -6.57712396 & -4.19505288 & -35512.33278793 \\ 37260.00257254 & -35512.37148912 & -0.75947834 \end{pmatrix}$$

Mode B_1

Frequency(THZ) 5.58993169

Raman tensor:

$$\begin{pmatrix} 1.80311736 & 5.30730249 & 40310.27243942 \\ 5.29499864 & 11.36345116 & -38418.17338309 \\ 40310.26375434 & -38418.18230941 & 1.05933748 \end{pmatrix}$$

Mode B_1

Frequency(THZ) 8.37149125

Raman tensor:

$$\begin{pmatrix} -4.20491631 & -3.16643756 & 18468.45914604 \\ -3.13260755 & -2.63976583 & -17603.18004260 \\ 18468.48246825 & -17603.15569524 & -0.31395273 \end{pmatrix}$$

Mode B_2

Frequency(THZ) 1.74441423

Raman tensor:

$$\begin{pmatrix} -1585.67500906 & 1515.37221242 & -0.63314223 \\ 1515.23933328 & -1440.17896644 & -2.19871523 \\ -0.57767243 & -2.24714782 & 3028.84189351 \end{pmatrix}$$

Mode B_2

Frequency(THZ) 2.03938203

Raman tensor:

$$\begin{pmatrix} -350.71364849 & 335.12478983 & 0.63775152 \\ 335.15663919 & -319.72371498 & 0.57201444 \\ 0.55163086 & 0.65074605 & 673.64217312 \end{pmatrix}$$

Mode B_2

Frequency(THZ) 4.10472914

Raman tensor:

$$\begin{pmatrix} -2625.56800471 & 2506.50737113 & -0.69657832 \\ 2506.45130381 & -2385.62622908 & -0.49091202 \\ -0.76659787 & -0.42657671 & 5013.41832361 \end{pmatrix}$$

Mode B_2

Frequency(THZ) 4.90229556

Raman tensor:

$$\begin{pmatrix} -1702.70902154 & 1931.72070111 & 1.38510468 \\ 1931.77031757 & -1516.19033759 & 0.25379731 \\ 1.37835060 & 0.25327777 & 3575.02842280 \end{pmatrix}$$

Mode B_2

Frequency(THZ) 4.93921317

Raman tensor:

$$\begin{pmatrix} -1464.61888112 & 1398.03140180 & -0.48798966 \\ 1398.05387333 & -1331.53304252 & -0.54646671 \\ -0.46398599 & -0.57762042 & 2797.38147374 \end{pmatrix}$$

Mode B_2

Frequency(THZ) 5.46881771

Raman tensor:

$$\begin{pmatrix} 1054.34891476 & -1006.94522475 & 0.36241315 \\ -1006.91322523 & 957.48413933 & 1.24103559 \\ 0.39466073 & 1.21846229 & -2013.72077763 \end{pmatrix}$$

Mode B_2

Frequency(THZ) 8.37679605

Raman tensor:

$$\begin{pmatrix} -1116.24304588 & 1064.27598951 & -0.13977084 \\ 1064.28445270 & -1014.25828761 & 0.51984496 \\ -0.15515846 & 0.53164213 & 2130.93064797 \end{pmatrix}$$

Mode E

Frequency(THZ) 1.70482819

Raman tensor:

$$\begin{pmatrix} -5061.96652679 & -242.48857403 & 442.52582641 \\ -242.22693578 & 5062.62785822 & 463.95512402 \\ 442.53823068 & 463.96592033 & 0.05696776 \end{pmatrix}$$

Mode E

Frequency(THZ) 1.70482872

Raman tensor:

$$\begin{pmatrix} -638.81980890 & -30.40861485 & -3500.46803856 \\ -30.28549096 & 639.83029770 & -3672.25297205 \\ -3500.61574129 & -3672.42020003 & 0.06799379 \end{pmatrix}$$

Mode E

Frequency(THZ) 1.82903138

Raman tensor:

$$\begin{pmatrix} 3142.87167889 & 150.38612867 & 15421.0060253 \\ 150.17802674 & -3143.48213962 & 16177.7939426 \\ 15421.86484518 & 16178.69791851 & -0.04366520 \end{pmatrix}$$

Mode E

Frequency(THZ) 1.82903185

Raman tensor:

$$\begin{pmatrix} -22330.41745491 & -1069.95725601 & 2171.21714 \\ -1068.74677159 & 22332.62670080 & 2277.621389 \\ 2171.34557181 & 2277.75871851 & 0.05601925 \end{pmatrix}$$

Mode E

Frequency(THZ) 2.06381497

Raman tensor:

$$\begin{pmatrix} 13606.03260133 & 652.29102466 & 174.90253989 \\ 651.09211517 & -13606.85509177 & 183.52080011 \\ 174.91164339 & 183.52905677 & -0.09018814 \end{pmatrix}$$

Mode E

Frequency(THZ) 2.06381594

Raman tensor:

$$\begin{pmatrix} -254.33312229 & -12.63713811 & 9404.23845981 \\ -12.57976490 & 253.80702475 & 9865.77525667 \\ 9405.23899782 & 9866.80077638 & -0.01820700 \end{pmatrix}$$

Mode E

Frequency(THZ) 2.84954306

Raman tensor:

$$\begin{pmatrix} -43.30405595 & -1.49312981 & 9724.82475710 \\ -1.47173557 & 44.89052107 & 10202.02373153 \\ 9725.37701059 & 10202.60560782 & 0.07828881 \end{pmatrix}$$

Mode E

Frequency(THZ) 2.84954393

Raman tensor:

$$\begin{pmatrix} -14083.59763231 & -676.07444280 & -30.32456147 \\ -675.30589614 & 14082.23898099 & -31.86306541 \\ -30.32127005 & -31.85812828 & -0.06629861 \end{pmatrix}$$

Mode E

Frequency(THZ) 3.14736076

Raman tensor:

$$\begin{pmatrix} 16787.17309279 & 805.06596441 & -1741.8521340 \\ 803.97685707 & -16787.51821744 & -1827.219828 \\ -1741.95415724 & -1827.32563826 & 0.01846346 \end{pmatrix}$$

Mode E

Frequency(THZ) 3.1473609

Raman tensor:

$$\begin{pmatrix} -2521.46282061 & -120.6711969 & -11591.11914519 \\ -120.49413714 & 2521.79516273 & -12160.01025818 \\ -11591.88798222 & -12160.8288045 & 0.02461793 \end{pmatrix}$$

Mode E

Frequency(THZ) 3.32832009

Raman tensor:

$$\begin{pmatrix} 1435.78177390 & 69.47119822 & 13208.27212402 \\ 69.43676828 & -1434.09668840 & 13856.62332441 \\ 13209.15714688 & 13857.54698807 & 0.07133686 \end{pmatrix}$$

Mode E

Frequency(THZ) 3.32832068

Raman tensor:

$$\begin{pmatrix} -19121.60343678 & -915.35950164 & 992.47190911 \\ -914.12126241 & 19125.79918282 & 1040.98790093 \\ 992.54076898 & 1041.066421 & 0.25488061 \end{pmatrix}$$

Mode E

Frequency(THZ) 4.12334932

Raman tensor:

$$\begin{pmatrix} 3179.91953357 & 152.52642837 & 14187.74355767 \\ 152.37153734 & -3179.59428818 & 14884.39959496 \\ 14188.74300427 & 14885.39053674 & -0.01958689 \end{pmatrix}$$

Mode E

Frequency(THZ) 4.12334952

Raman tensor:

$$\begin{pmatrix} 20553.05741704 & 985.67084287 & -2195.83857882 \\ 984.48171273 & -20553.27983128 & -2303.88989083 \\ -2195.97233663 & -2304.02699903 & 0.02319500 \end{pmatrix}$$

Mode E

Frequency(THZ) 4.41219556

Raman tensor:

$$\begin{pmatrix} 12439.73392422 & 597.30153599 & -451.52943206 \\ 596.23430393 & -12438.38449386 & -473.63427555 \\ -451.58064678 & -473.68005841 & 0.06776898 \end{pmatrix}$$

Mode E

Frequency(THZ) 4.41219563

Raman tensor:

$$\begin{pmatrix} 651.33099946 & 32.31235164 & 8598.24326142 \\ 32.32114609 & -648.37761710 & 9019.90416357 \\ 8598.97138993 & 9020.61418586 & 0.17200904 \end{pmatrix}$$

Mode E

Frequency(THZ) 4.63984006

Raman tensor:

$$\begin{pmatrix} -14940.07992921 & -718.20646405 & 36631.21460632 \\ -717.22954151 & 14936.10554360 & 38429.43282737 \\ 36633.80525259 & 38432.14127674 & -0.26371542 \end{pmatrix}$$

Mode E

Frequency(THZ) 4.63984038

Raman tensor:

$$\begin{pmatrix} 53050.4945242 & 2544.30185835 & 10313.42232615 \\ 2540.77236474 & -53051.21463086 & 10819.85022970 \\ 10314.15239879 & 10820.59895662 & -0.02504274 \end{pmatrix}$$

Mode E

Frequency(THZ) 5.28506058

Raman tensor:

$$\begin{pmatrix} -26506.84199491 & -1271.36218807 & -1352.52503133 \\ -1269.58971554 & 26506.93088674 & -1419.12596155 \\ -1352.62643954 & -1419.21970668 & 0.05338619 \end{pmatrix}$$

Mode E

Frequency(THZ) 5.28506125

Raman tensor:

$$\begin{pmatrix} 1948.97800529 & 84.08452374 & -18310.36165292 \\ 83.96523498 & -1971.27733951 & -19208.19115442 \\ -18311.64981822 & -19209.54139074 & -1.14818620 \end{pmatrix}$$

Mode E

Frequency(THZ) 5.83438864

Raman tensor:

$$\begin{pmatrix} -6737.04529753 & -298.68871502 & -22.00528621 \\ -298.92101411 & 6792.88631982 & -25.41211244 \\ -22.03707690 & -25.44072405 & 2.69539607 \end{pmatrix}$$

Mode E

Frequency(THZ) 5.83438898

Raman tensor:

$$\begin{pmatrix} 32.07521643 & -0.72119450 & -4659.42174592 \\ -0.60697510 & -35.78360039 & -4888.233873 \\ -4659.25121158 & -4888.07037802 & -0.06562507 \end{pmatrix}$$

Mode E

Frequency(THZ) 5.99848998

Raman tensor:

$$\begin{pmatrix} 52064.45450806 & 52064.45450806 & -2053.02917513 \\ 2494.32223487 & -52062.95837287 & -2153.29849485 \\ -2053.18034416 & -2153.45820821 & 0.04096900 \end{pmatrix}$$

Mode E

Frequency(THZ) 5.99849037

Raman tensor:

$$\begin{pmatrix} 2970.52513615 & 142.34773888 & 35951.27982838 \\ 142.12733886 & -2970.81038547 & 37715.71012787 \\ 35953.89132758 & 37718.45132569 & 0.01117336 \end{pmatrix}$$

Mode E

Frequency(THZ) 6.51025892

Raman tensor:

$$\begin{pmatrix} 9978.27897519 & 480.74494787 & -86.25095186 \\ 480.27405415 & -9973.30492852 & -90.52028013 \\ -86.29382358 & -90.56152967 & 0.27901380 \end{pmatrix}$$

Mode E

Frequency(THZ) 6.51025947

Raman tensor:

$$\begin{pmatrix} 125.03363826 & 5.55177209 & 6897.85075901 \\ 5.6319538 & -125.96940048 & 7236.28615522 \\ 6898.29732013 & 7236.72321504 & -0.10822213 \end{pmatrix}$$

Mode E

Frequency(THZ) 7.02437468

Raman tensor:

$$\begin{pmatrix} -2454.11595827 & -113.88646253 & 8175.83573923 \\ -113.66208538 & 2463.55270441 & 8576.57831842 \\ 8176.39611608 & 8577.15861978 & 0.49584861 \end{pmatrix}$$

Mode E

Frequency(THZ) 7.02437499

Raman tensor:

$$\begin{pmatrix} 11829.75521083 & 567.12401287 & 1703.19648979 \\ 566.37707016 & -11829.93611633 & 1786.68494222 \\ 1703.30335762 & 1786.78750816 & -0.01833961 \end{pmatrix}$$

Mode E

Frequency(THZ) 8.29654544

Raman tensor:

$$\begin{pmatrix} 44022.57501345 & 2111.17402838 & 439.02688076 \\ 2108.18415469 & -44023.75505452 & 460.8772744 \\ 439.04928342 & 460.90647337 & -0.13642966 \end{pmatrix}$$

Mode E

Frequency(THZ) 8.29654627

Raman tensor:

$$\begin{pmatrix} -638.17359344 & -31.30557001 & 30406.4100218 \\ -31.21193195 & 636.84277508 & 31898.85925464 \\ 30408.55111145 & 31901.10958869 & -0.0297024 \end{pmatrix}$$

Mode E

Frequency(THZ) 8.61367196

Raman tensor:

$$\begin{pmatrix} -62568.34388859 & -3001.71247209 & 3484.95396965 \\ -2996.87283498 & 62568.07425587 & 3656.18891545 \\ 3485.27811826 & 3656.51969438 & 0.01424289 \end{pmatrix}$$

Mode E

Frequency(THZ) 8.61367246

Raman tensor:

$$\begin{pmatrix} 5041.18432184 & 239.77246223 & 43230.49463558 \\ 239.42817714 & -5045.55197844 & 45352.20330044 \\ 43233.95516729 & 45355.86740727 & -0.27847310 \end{pmatrix}$$

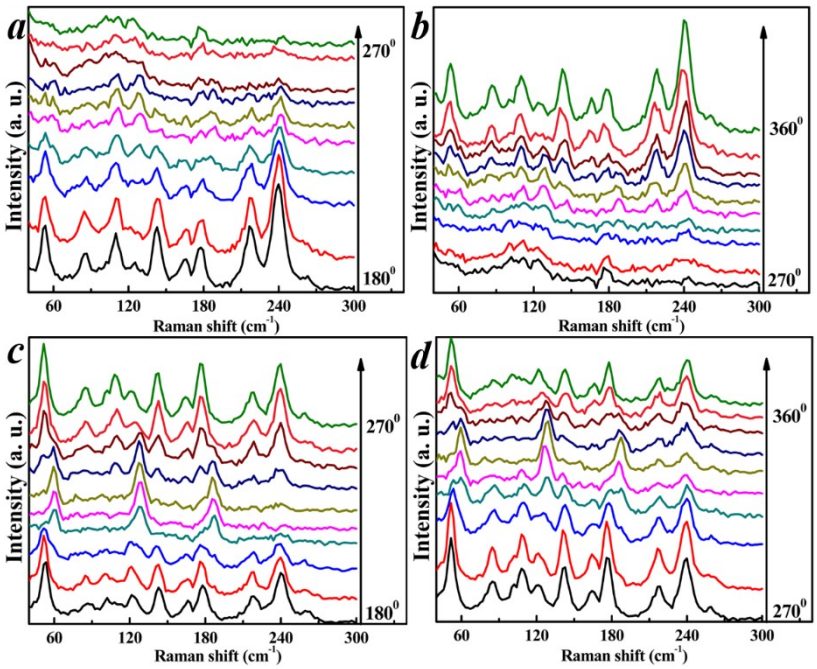

Fig. 1 The relationship between Raman scattering intensity and the rotating angle of the sample for (110) surface from 180° to 360° in 10° steps, (a) and (b) parallel polarization, (c) and (d) vertical polarization

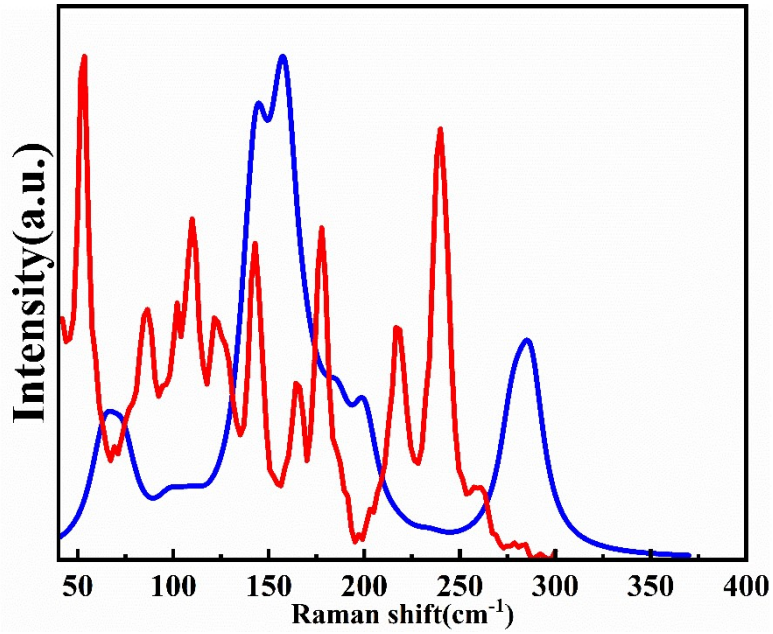

Fig. 2 The red line represents the normalized Raman spectrum of the (110) surface of layered $(TaSe_4)_2I$ single crystal, the blue line represents the result of the density functional theory calculation.