Supplementary Information for "Revealing the role of ripples in phonon modes for MoS_2 and $MoSe_2$: Insights from molecular dynamics and machine learning"

4.1 Convergence of DFT Calculations

Our DFT Calculations for MoS_2 and $MoSe_2$ were properly converged for both the Plane Wave Kinetic Energy and **k**-points criterion. The energy convergence criterion is shown below. Since we are using Norm-Conserved pseudopotentials, the selected energy cutoff for charge density is always 4 times the cutoff energy for wavefunctions. We conducted convergence tests, which are presented below. Figure S1 shows the energy as a function of the plane-wave cutoff energy for three different k-point grids: $3\times3\times1$, $6\times6\times1$, and $24\times24\times1$. Notably, the energy curves for the $6\times6\times1$ grid overlap with those for the $24\times24\times1$ grid, whereas the $3\times3\times1$ grid shows small deviations. It is noticeable that $6\times6\times1$ and $24\times24\times1$ grid results indicate similar accuracy, within negligible deviations. Figure S2 presents the difference in energy per atom between the $6\times6\times1$ and $24\times24\times1$ grids as a function of cutoff energy. For cutoff values of 40 Ry or higher, the difference stabilizes at approximately 1.4 meV/atom. These results suggest that, for both MoS_2 and $MoSe_2$, increasing the k-point grid beyond $6\times6\times1$ does not improve energy convergence, particularly when using a plane wave cutoff of at least 40 Ry. The similarity between the $6\times6\times1$ and $24\times24\times1$ results supports the adequacy of our chosen parameters.

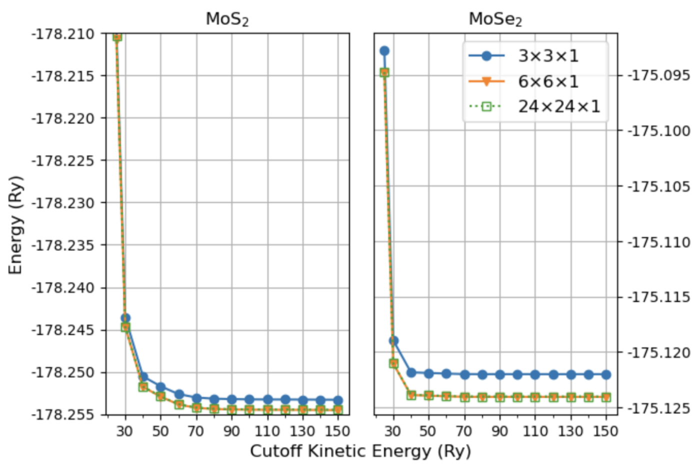

Figure S1 Energy convergence criterion for the plane wave expansion of wavefunctions for several k-points grid.

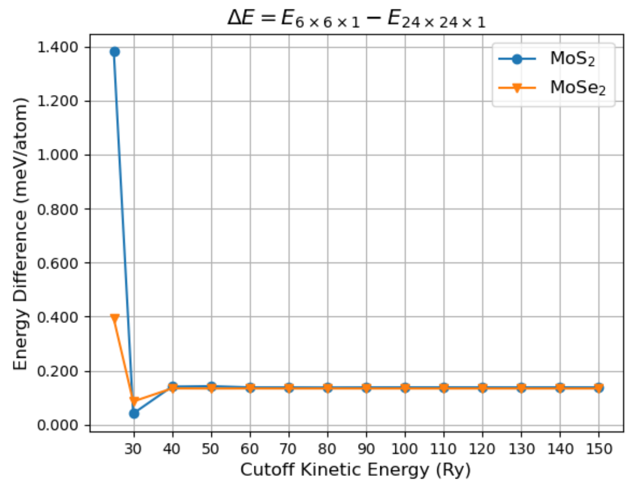

Figure S2 Energy difference for the $6\times6\times1$ and $24\times24\times1$ convergence curves.

As further validation we directly compared energy value per atom results using our chosen cutoff energies, namely 70 Ry for MoS_2 and 40 Ry for $MoSe_2$ [both with a grid of ($6\times6\times1$)], with results from a higher plane-wave cutoff (150 Ry) and a denser k-point sampling ($24\times24\times1$). The resulting differences in energy per atom are 1.068 meV for MoS_2 and 0.618 meV for $MoSe_2$. These values fall within the typical 1–2 meV/atom energy convergence criterion. This choice of parameters allowed that we could generate a DFT dataset of thousand of structures without sacrificing the accuracy and convergence of each DFT structure.

4.2 Accuracy and Validation of MLFF

To check the accuracy of the MLFFs generated in this work on the task of predicting the energies and atomic forces on unseen data we have included Figures S3 and S4. Each one of the subfigures was made with 750 structures of the testing set generated at the time of splitting the reference set between training and testing data. Figure S3a) and b) shows a comparison between the energies per atom and the absolute value for the atomic forces predicted by the MLFF and the reference DFT values on the data set respectively for MoS_2, with the dashed black line indicating perfect accuracy for the MLFF, i.e., when the MLFF matches DFT without any error. For the structures included in the figure, we obtained a RMSE of 4.1 meV/atom and 286.16 meV/Å for the energies and forces respectively. Figure S3 c) shows a histogram of angles between the direction of the MLFF and DFT forces which is a measure of directional error in the sense that the more accurate an MLFF is, the more concentrated at 0° the distribution becomes. Due to the tail of the distribution on angles ≥10%, we fitted a Lorentz distribution to the histogram. The red dashed line indicates the fitted curve which has a full width half maximum (FWHM) equal to 6.47° and, of all the forces evaluated with the MLFF and included in the histogram, 60.2% of them have their angle with the DFT force within the range of the FWHM and 72.4% of them have this angle within the range of 10° (see inset at Figure S3). The accuracy check of the MLFF for $MoSe_2$ in Figure S4 with each one of the subfigures being made with the same amount structures of the testing set as in Figure S3. Figure S4a) and b) shows a comparison between the energies per atom and the absolute value for the atomic forces predicted by the MLFF and the reference DFT values on the data set respectively. For the structures included in the figure, we obtained a RMSE of 1.69 meV/atom and 184.11 meV/Å for the energies and forces respectively, both being slightly more accurate than the RMSE obtained for MoS_2. Figure S4 c) shows a histogram of angles between the direction of the MLFF and DFT forces. The red dashed line indicates the Lorentzian fitted to the histogram with a FWHM = 7.23° and, of all the forces evaluated with the MLFF and included in the histogram, 64.7% of them have their angle with the DFT force within the FWHM and 72.1% of them have this angle within the range of 10°. Both figures indicates good agreement between the MLFF and DFT estimates for the force, especially on predicting the direction.

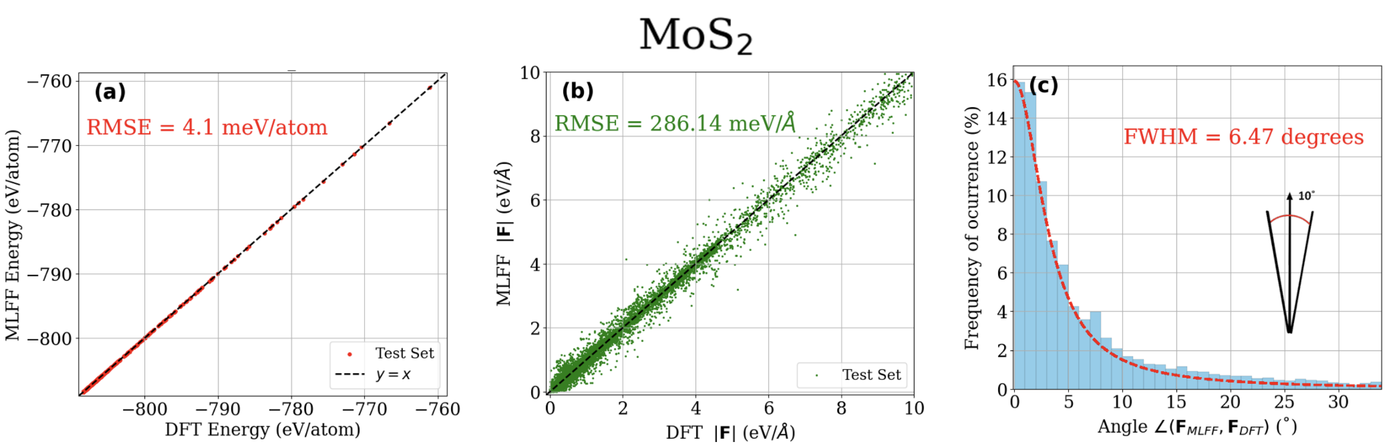

Figure S3 (a,b) Comparison of the energies and lengths of the atomic forces between MLFF and DFT for MoS_2 based respectively.(c) Histogram of the directional errors in the MLFF prediction of the forces. The red dashed line indicates the fitted Lorentzian function.

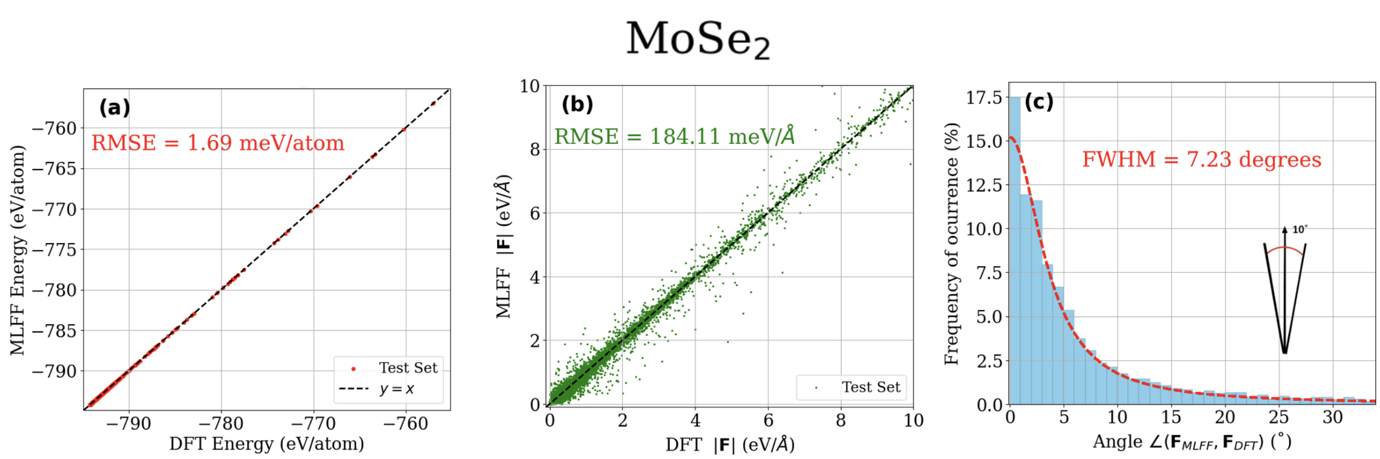

Figure S4 (a,b) Comparison of the energies and lengths of the atomic forces between MLFF and DFT for $MoSe_2$ based respectively.(c) Histogram of the directional errors in the MLFF prediction of the forces. The red dashed line indicates the fitted Lorentzian function.

The comparison of the MLFF and DFT forces by each component is included here as a complement to the comparison of the force modules and the histogram shown in Figures S3 and S4.

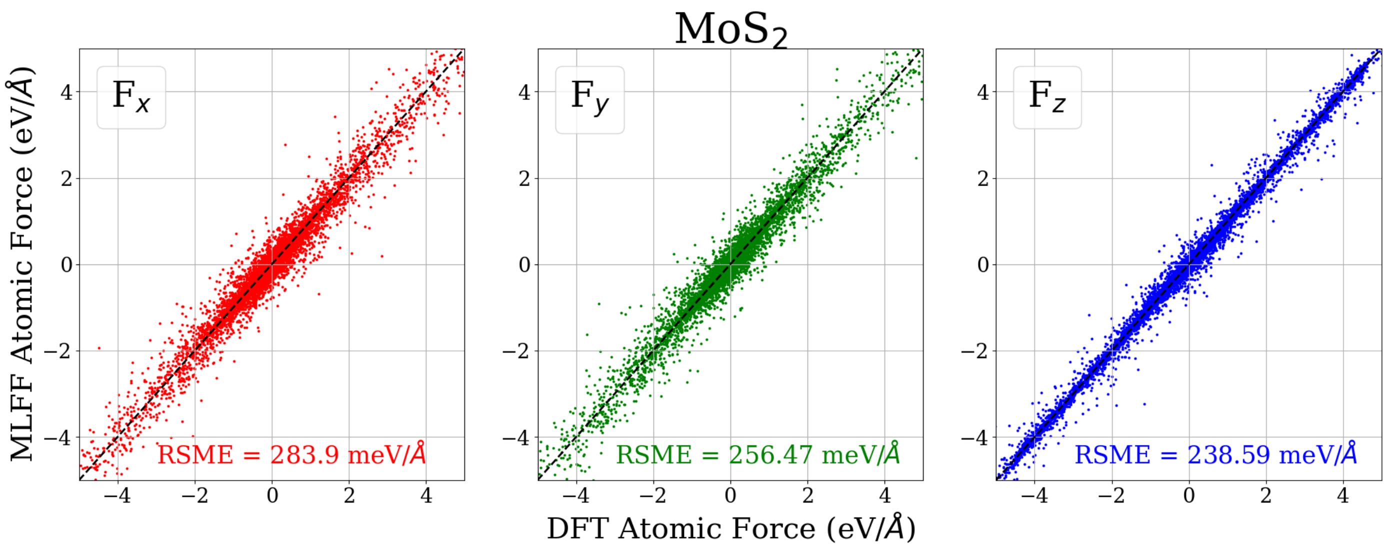

Figure S5 Comparison of the atomic forces Cartesian components between the MLFF developed for MoS_2 and the DFT reference set based on structures included in the testing set exclusively.

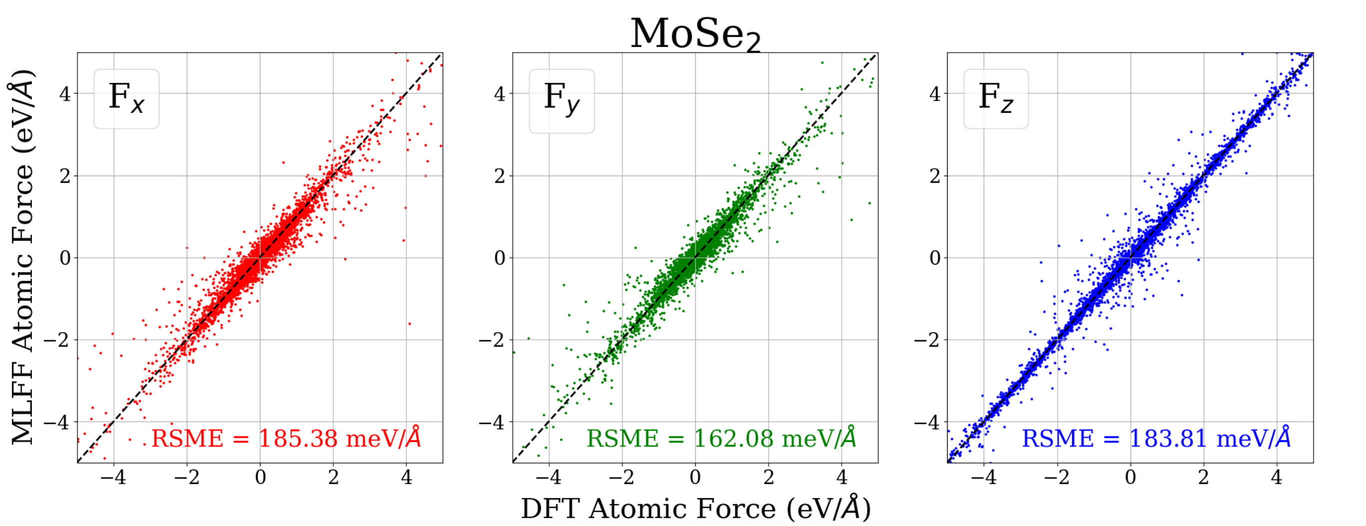

Figure S6 Comparison of the atomic forces Cartesian components between the MLFF developed for $MoSe_2$ and the DFT reference set based on structures included in the testing set exclusively.

4.3 Dielectric Spectra in Semi-log Plot

Figure S7 shows the imaginary part of dielectric spectra for MoS_2 and $MoSe_2$ in a semi-log scale, which is more suitable for comparing relative intensities of the observed peaks. Here, since the y axis is in log scale, we have omitted the errorbars for the sake of clarity of the figure.

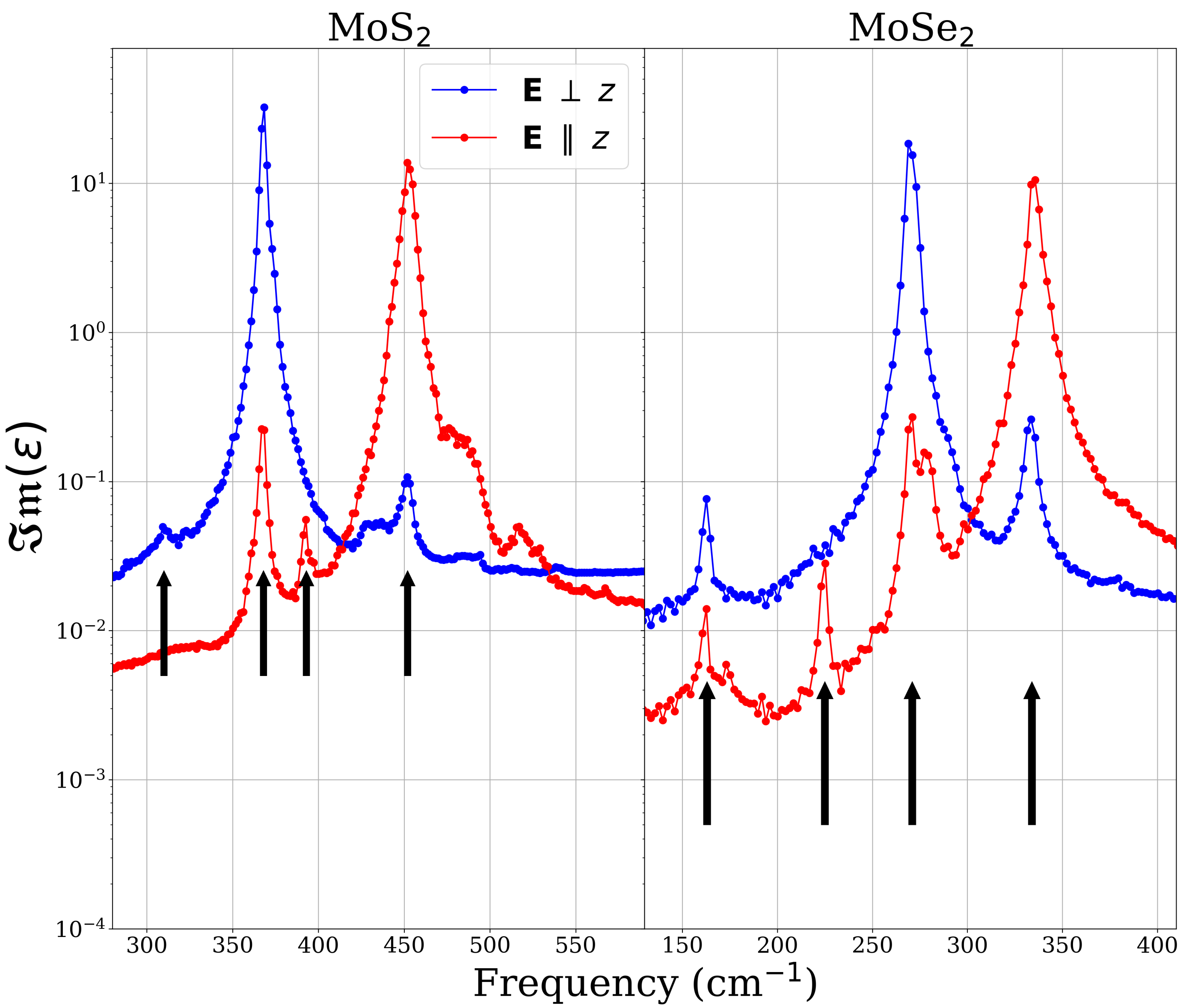

Figure S7 Imaginary part of dielectric spectra for MoS_2 and $MoSe_2$ at 300K in semi-log scale. Arrows indicate peaks centered at 310(163), 368(225), 393(271) and 452(334) for MoS_2 ($MoSe_2$) in cm^{-1}.

4.4 Effect of Tensile Strain on Dielectric Spectra

Figure S8 shows the newly identified peaks in the imaginary part of the dielectric spectra for unstrained and strained lattices (1.4% strain relative to the lattice constant). All simulations were performed with constant volume (NVT ensemble) throughout the dynamics which allowed us to assign a strain value to each curve shown. By observing each one of the peaks, it is evident that the application of strain flattens all newly observed peaks, thereby extinguishing the phenomenon observed in the unstrained lattice.

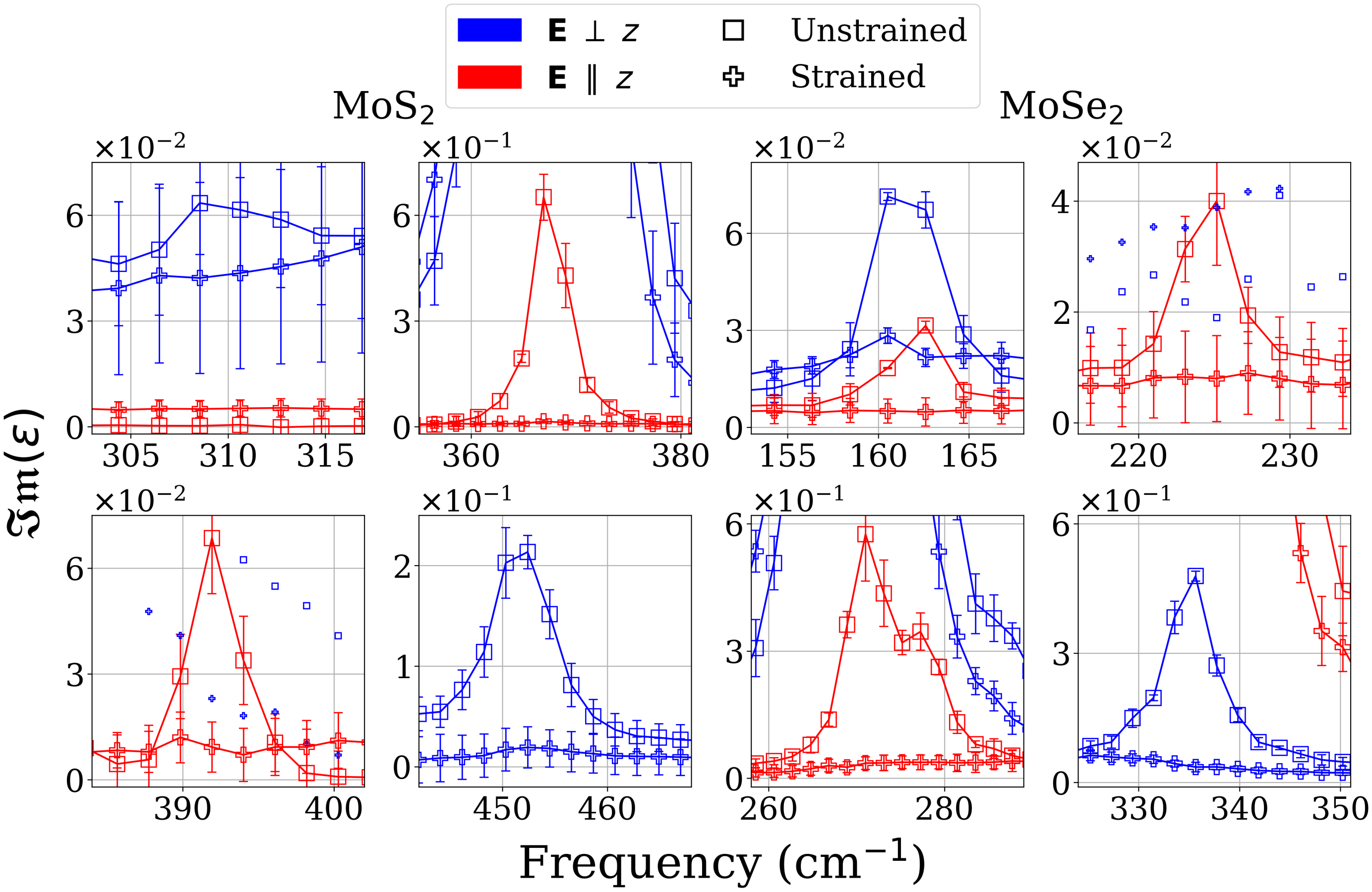

Figure S8 All found low intensity peaks under the effect of strain for MoS_2 and $MoSe_2$. In some graphics, blue markers were reduced with the purpose of avoiding visual overload.

4.5 Calculation of Eigenmodes on Rippled Supercell

The supercell used on the calculation of eigenmodes is included in Figure S9 and was used for both MoS_2 and $MoSe_2$. This cell was constructed by the replication of the rectangular unit cell of $MoS(Se)_2$ (see green rectangle at Figure 1 at main manuscript). The unit cell was replicated 17 times in the direction of $\mathbf{a}_1$ and 10 times in the direction of $\mathbf{a}_2$, resulting on a approximately square cell with a total of 1020 atoms. In order to model the ripple, an overall sine function with wave vector parallel to the zig-zag direction (referred as $\mathbf{x}$ in the main manuscript) and peak-to-peak amplitude of 1Å were added to the z component of each atom of the supercell:

$$\Delta z_i = (0.5\ \text{Å}) \times \sin\left(\frac{2\pi}{17a_0} x_i\right).$$

The amplitude was chosen so that the deformed system is very similar to the original, but the perturbation allows a qualitative investigation of any new peaks in the spectra without losing correspondence of peaks between the perturbed and original systems. After adding the deformation, the energy of the model cell must be minimized to ensure that the dynamical matrix is being calculated at a local minimum of the energy. Minimization usually does not significantly affect the model structure and can eliminate strain in the $\mathbf{x}$ direction. After minimization, the dimensions for each cell were 54.26 Å× 55.28 Å for MoS_2 and 56.46 Å× 57.53 Å for $MoSe_2$. Alamode detected that this supercell can be assigned to a primitive cell with 102 atoms shown in Figure S10. Therefore, the calculation resulted in a total of 306 eigenmodes, some of which were mentioned in the main manuscript.

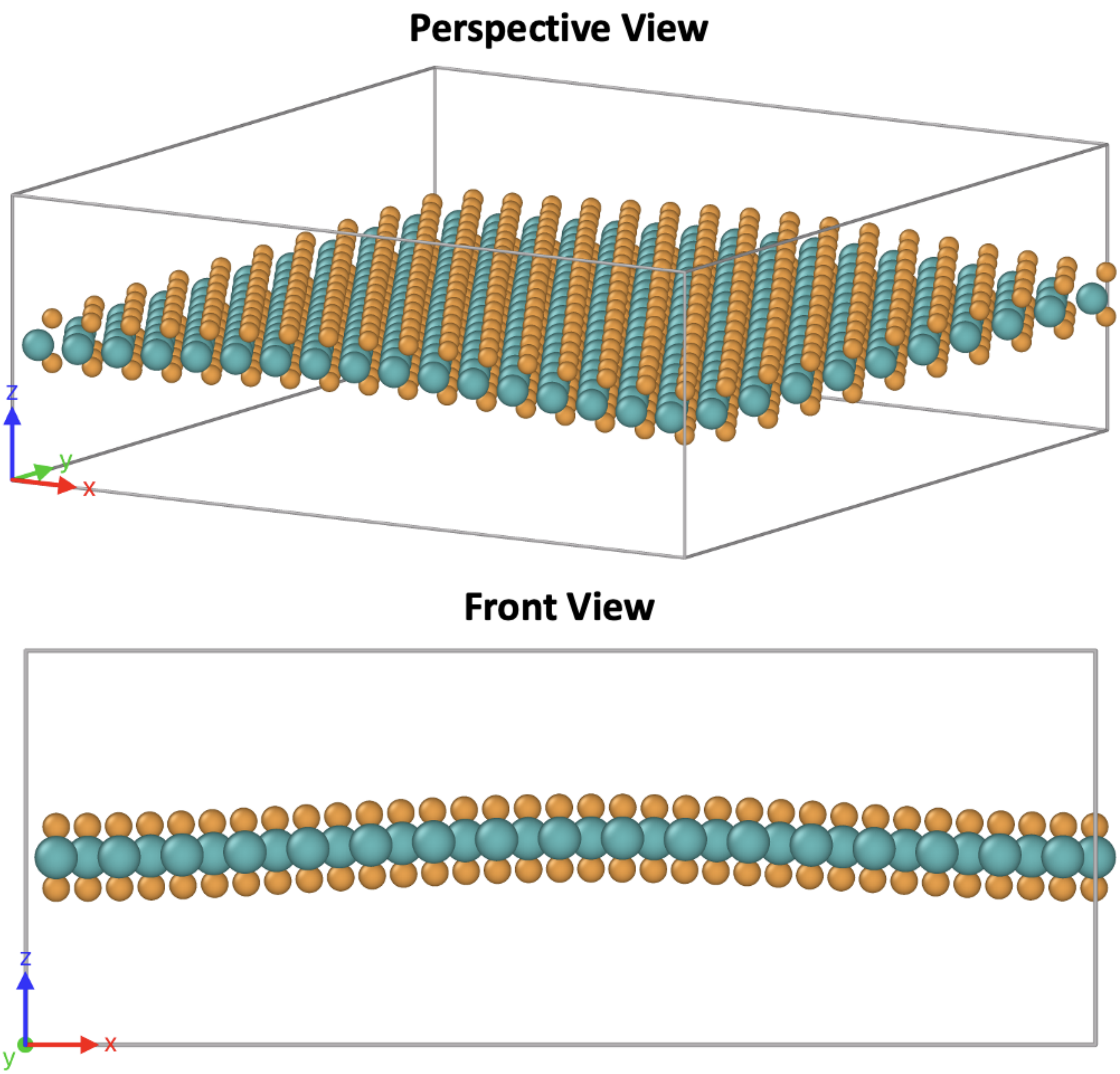

Figure S9 Supercell of 1020 atoms containing a sinusoidal wave in the x direction used in the calculation made by finite displacements.

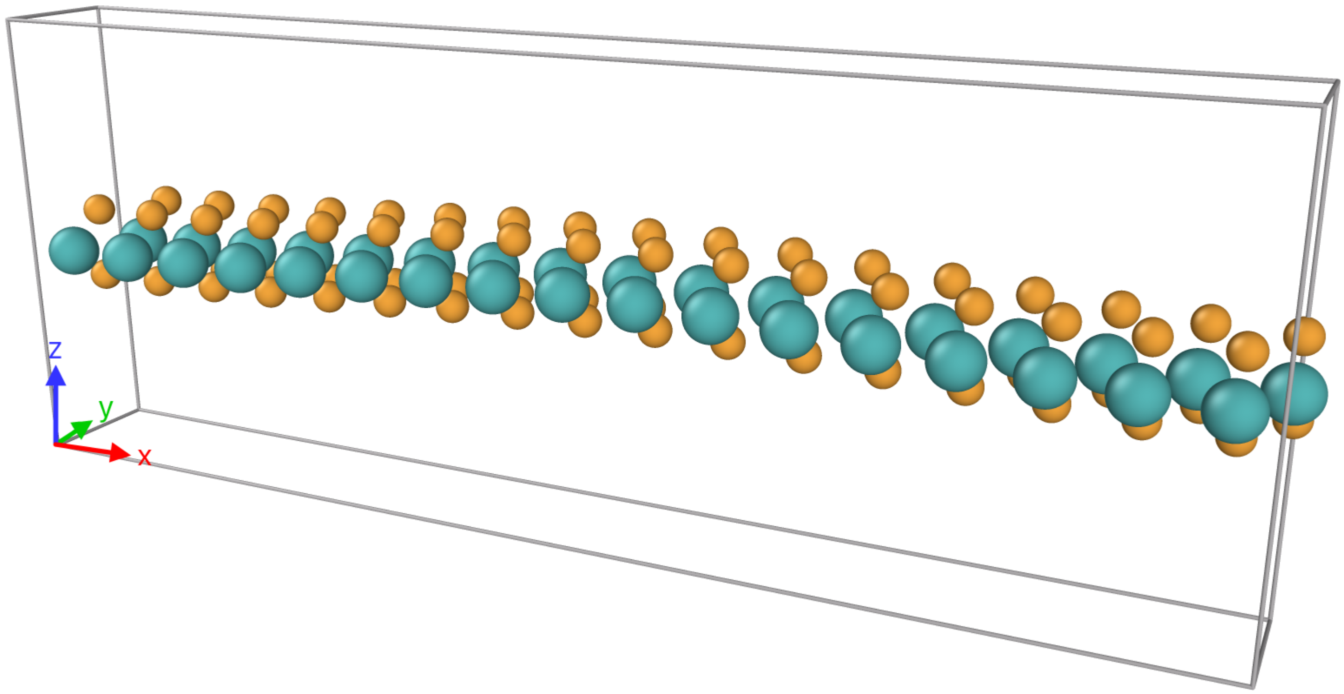

Figure S10 Primitive rippled cell of 102 atoms. The supercell at Figure S9 is made by 10 units of this primitive cell repeated on the y (armchair) direction

The calculated frequencies and IR intensities (see Figures 20 and 21 in the main manuscript) for the modes of these models are listed in the following tables for both compounds:

Table S1 Frequencies (in cm^{-1}) and IR intensities (in arb. units) calculated for the MoS_2 rippled supercell (continued on next page)

#	Frequency	$f_{xy}(\omega_j)$	$f_z(\omega_j)$
1	0.0000	0.0000e+00	0.0000e+00
2	0.0000	0.0000e+00	0.0000e+00
3	0.0000	0.0000e+00	0.0000e+00
4	0.3004	1.5000e-09	0.0000e+00
5	1.8919	2.9843e-06	0.0000e+00
6	9.7114	0.0000e+00	0.0000e+00
7	9.7339	0.0000e+00	1.9521e-09
8	28.1362	0.0000e+00	0.0000e+00
9	28.2301	0.0000e+00	1.3760e-06
10	30.3155	7.8702e-12	0.0000e+00
11	30.3498	4.6923e-11	0.0000e+00
12	41.9107	0.0000e+00	1.2716e-06
13	41.9859	0.0000e+00	0.0000e+00
14	53.1230	3.3736e-07	0.0000e+00
15	53.2314	2.6503e-08	0.0000e+00
16	60.3363	0.0000e+00	0.0000e+00
17	60.3553	0.0000e+00	7.9234e-10
18	73.1519	0.0000e+00	0.0000e+00
19	73.1689	0.0000e+00	0.0000e+00
20	81.7425	2.3890e-07	0.0000e+00
21	81.8700	2.5765e-07	0.0000e+00
22	82.8899	1.6798e-09	0.0000e+00
23	82.9214	7.7406e-10	0.0000e+00
24	88.2026	0.0000e+00	0.0000e+00
25	88.2153	0.0000e+00	0.0000e+00
26	93.7257	5.9053e-10	0.0000e+00
27	93.7349	9.3762e-11	0.0000e+00
28	118.3505	1.2801e-10	0.0000e+00
29	118.3634	4.6785e-11	0.0000e+00
30	122.0125	0.0000e+00	2.7460e-09
31	122.0369	0.0000e+00	0.0000e+00
32	124.4398	0.0000e+00	7.7146e-11
33	124.4455	0.0000e+00	0.0000e+00
34	139.7821	1.9723e-11	0.0000e+00
35	139.7829	0.0000e+00	0.0000e+00
36	141.1141	0.0000e+00	0.0000e+00
37	141.1176	0.0000e+00	0.0000e+00
38	146.7319	0.0000e+00	0.0000e+00
39	146.7371	0.0000e+00	0.0000e+00
40	146.7722	0.0000e+00	0.0000e+00
41	147.4916	0.0000e+00	0.0000e+00
42	147.4994	2.3797e-11	0.0000e+00
43	148.8470	0.0000e+00	0.0000e+00
44	148.8540	0.0000e+00	0.0000e+00
45	150.4327	0.0000e+00	3.2671e-11
46	150.4336	0.0000e+00	0.0000e+00
47	156.3055	0.0000e+00	0.0000e+00
48	156.3070	3.4823e-11	0.0000e+00
49	157.6091	3.6204e-11	0.0000e+00
50	157.7530	2.4539e-11	0.0000e+00
51	158.3782	0.0000e+00	0.0000e+00
52	158.3796	0.0000e+00	0.0000e+00
53	159.1099	0.0000e+00	0.0000e+00
54	159.1399	0.0000e+00	0.0000e+00
55	161.6504	0.0000e+00	0.0000e+00
56	161.6510	0.0000e+00	0.0000e+00
57	162.7385	5.4753e-11	0.0000e+00
58	162.7441	0.0000e+00	0.0000e+00
59	163.0493	2.1695e-09	0.0000e+00
60	163.0657	3.4835e-09	0.0000e+00
61	164.8681	2.2028e-11	0.0000e+00
62	164.8754	2.0751e-11	0.0000e+00
63	168.7493	0.0000e+00	0.0000e+00
64	168.7521	4.4227e-11	0.0000e+00
65	172.0617	0.0000e+00	0.0000e+00
66	172.0736	0.0000e+00	0.0000e+00
67	174.9430	0.0000e+00	0.0000e+00
68	174.9444	0.0000e+00	9.1010e-12
69	175.0726	5.7227e-11	0.0000e+00
70	175.0895	2.1084e-11	0.0000e+00
71	175.1134	0.0000e+00	0.0000e+00
72	175.2960	0.0000e+00	0.0000e+00
73	177.3062	9.9039e-11	0.0000e+00
74	177.3329	4.8577e-11	0.0000e+00
75	177.3987	1.1791e-11	0.0000e+00
76	178.1685	0.0000e+00	0.0000e+00
77	178.1703	0.0000e+00	0.0000e+00
78	194.6444	0.0000e+00	0.0000e+00
79	194.6508	0.0000e+00	0.0000e+00
80	207.1886	0.0000e+00	0.0000e+00
81	207.2135	0.0000e+00	0.0000e+00
82	210.7835	6.2832e-11	0.0000e+00
83	210.7954	1.6482e-11	0.0000e+00
84	211.9602	1.7368e-09	0.0000e+00
85	211.9632	4.9071e-10	0.0000e+00
86	214.3197	0.0000e+00	0.0000e+00
87	214.3218	0.0000e+00	0.0000e+00
88	218.6117	0.0000e+00	0.0000e+00
89	218.6140	2.6231e-11	0.0000e+00
90	222.7617	0.0000e+00	0.0000e+00
91	222.7675	0.0000e+00	0.0000e+00
92	227.7333	0.0000e+00	0.0000e+00
93	227.7431	0.0000e+00	0.0000e+00
94	229.2407	0.0000e+00	0.0000e+00
95	229.2421	0.0000e+00	0.0000e+00
96	229.2629	0.0000e+00	0.0000e+00
97	229.3726	1.1092e-11	0.0000e+00
98	229.6204	0.0000e+00	1.2724e-11
99	232.0023	0.0000e+00	0.0000e+00
100	232.0219	0.0000e+00	0.0000e+00
101	236.0732	0.0000e+00	0.0000e+00
102	236.1080	2.1504e-11	0.0000e+00
103	273.2504	0.0000e+00	2.7727e-06
104	273.2724	0.0000e+00	0.0000e+00
105	273.4864	1.6840e-04	0.0000e+00
106	273.5141	1.4075e-05	0.0000e+00
107 *E"(x)	273.7792	4.4171e-04	0.0000e+00
108	273.8270	4.8997e-05	0.0000e+00
109	274.0492	0.0000e+00	3.6610e-06
110	274.4042	0.0000e+00	0.0000e+00
111	275.3041	0.0000e+00	1.4193e-08
112	275.3100	0.0000e+00	0.0000e+00
113	276.5469	1.5850e-05	0.0000e+00
114 *E"(y)	276.8662	7.4914e-04	0.0000e+00
115	278.5667	4.0210e-10	0.0000e+00
116	278.5677	0.0000e+00	0.0000e+00
117	282.8495	0.0000e+00	1.0258e-09
118	282.8622	0.0000e+00	0.0000e+00
119	283.5313	0.0000e+00	0.0000e+00
120	283.5965	0.0000e+00	5.9290e-08
121	287.6733	1.5370e-08	0.0000e+00
122	287.6948	2.0589e-10	0.0000e+00
123	291.9335	5.2952e-10	0.0000e+00
124	291.9499	2.0494e-10	0.0000e+00
125	292.2277	0.0000e+00	3.3894e-10
126	292.2542	0.0000e+00	0.0000e+00
127	292.3891	0.0000e+00	0.0000e+00
128	292.4098	0.0000e+00	2.2970e-11
129	293.7746	0.0000e+00	0.0000e+00
130	293.7807	0.0000e+00	1.3956e-10
131	293.9443	1.2764e-08	0.0000e+00
132	293.9559	2.0135e-09	0.0000e+00
133	294.7139	1.6856e-11	0.0000e+00
134	294.7309	1.4578e-10	0.0000e+00
135	294.9363	1.4523e-10	0.0000e+00
136	294.9548	1.4455e-09	0.0000e+00
137	297.5955	0.0000e+00	0.0000e+00
138	297.6363	0.0000e+00	0.0000e+00
139	300.1429	2.2061e-10	0.0000e+00
140	300.2090	3.7842e-10	0.0000e+00
141	302.9717	0.0000e+00	5.5150e-10
142	303.0660	0.0000e+00	0.0000e+00
143	304.5511	5.2804e-11	0.0000e+00
144	306.0252	0.0000e+00	8.7459e-11
145	306.0271	0.0000e+00	0.0000e+00
146	316.7751	0.0000e+00	9.8539e-11
147	316.7796	0.0000e+00	0.0000e+00
148	318.3125	5.5392e-10	0.0000e+00
149	318.3265	1.2964e-11	0.0000e+00
150	320.8214	1.3012e-09	0.0000e+00
151	321.6145	0.0000e+00	0.0000e+00
152	321.8978	0.0000e+00	1.2098e-10

#	Frequency	$f_{xy}(\omega_j)$	$f_z(\omega_j)$
153	323.7785	2.9885e-09	0.0000e+00
154	323.7843	2.1762e-09	0.0000e+00
155	323.8573	1.2087e-09	0.0000e+00
156	323.9123	1.5143e-10	0.0000e+00
157	326.3453	0.0000e+00	3.9333e-11
158	326.3629	0.0000e+00	0.0000e+00
159	328.0523	1.7098e-09	0.0000e+00
160	328.0574	4.3551e-10	0.0000e+00
161	328.2593	0.0000e+00	2.9334e-11
162	328.2600	0.0000e+00	0.0000e+00
163	328.9067	2.8267e-10	0.0000e+00
164	328.9304	1.0511e-10	0.0000e+00
165	329.1346	0.0000e+00	1.2178e-09
166	329.1574	0.0000e+00	0.0000e+00
167	329.1876	3.8037e-10	0.0000e+00
168	329.2029	9.0030e-10	0.0000e+00
169	329.2486	0.0000e+00	1.9500e-09
170	329.2655	0.0000e+00	0.0000e+00
171	331.8733	6.7552e-11	0.0000e+00
172	331.8759	1.5611e-09	0.0000e+00
173	333.1575	0.0000e+00	0.0000e+00
174	333.1576	0.0000e+00	8.3908e-12
175	335.7557	2.1390e-09	0.0000e+00
176	335.7563	2.1324e-09	0.0000e+00
177	339.8761	0.0000e+00	0.0000e+00
178	339.8789	0.0000e+00	3.7071e-10
179	343.8019	0.0000e+00	0.0000e+00
180	343.8043	0.0000e+00	7.6687e-11
181	344.9890	3.2593e-10	0.0000e+00
182	344.9989	1.3071e-09	0.0000e+00
183	347.3096	4.6152e-10	0.0000e+00
184	347.3193	4.9868e-10	0.0000e+00
185	347.4101	0.0000e+00	2.1975e-11
186	347.4224	0.0000e+00	0.0000e+00
187	347.5440	0.0000e+00	4.5060e-11
188	347.5689	0.0000e+00	0.0000e+00
189	348.9126	1.7356e-09	0.0000e+00
190	348.9159	1.5788e-10	0.0000e+00
191	349.6935	1.3006e-08	0.0000e+00
192	349.6964	3.6542e-11	0.0000e+00
193	350.7227	0.0000e+00	5.7332e-10
194	350.7237	0.0000e+00	0.0000e+00
195	351.2465	0.0000e+00	0.0000e+00
196	351.2995	0.0000e+00	1.2422e-11
197	352.4320	0.0000e+00	0.0000e+00
198	352.4344	0.0000e+00	1.1743e-09
199	353.2188	8.9671e-10	0.0000e+00
200	353.6687	1.7246e-08	0.0000e+00
201	354.2405	0.0000e+00	0.0000e+00
202	354.8236	2.7183e-08	0.0000e+00
203	354.8381	1.7413e-07	0.0000e+00
204	354.9018	0.0000e+00	0.0000e+00
205	356.6375	3.7176e-08	0.0000e+00
206	357.0515	5.0216e-09	0.0000e+00
207	357.3550	5.9509e-09	0.0000e+00
208	357.3573	1.9804e-08	0.0000e+00
209	358.3473	9.9374e-09	0.0000e+00
210	358.3497	2.9031e-10	0.0000e+00
211	358.5896	0.0000e+00	3.2640e-10
212	358.5918	0.0000e+00	0.0000e+00
213	360.3862	0.0000e+00	0.0000e+00
214	360.3869	0.0000e+00	9.7288e-10
215	360.5574	1.9365e-07	0.0000e+00
216	360.5581	1.3874e-10	0.0000e+00
217	360.5696	1.1635e-08	0.0000e+00
218	360.5861	2.8598e-08	0.0000e+00
219	361.8122	0.0000e+00	3.6359e-07
220	361.8282	0.0000e+00	0.0000e+00
221	361.9944	0.0000e+00	1.9516e-09
222	362.0135	0.0000e+00	0.0000e+00
223	365.4927	9.4691e-10	0.0000e+00
224	365.5025	2.9297e-08	0.0000e+00
225	365.8672	4.2517e-06	0.0000e+00
226	365.8877	1.9735e-05	0.0000e+00
227	365.9394	0.0000e+00	3.6662e-10
228	366.0330	0.0000e+00	0.0000e+00
229	367.4811	0.0000e+00	6.5958e-06

#	Frequency	$f_{xy}(\omega_j)$	$f_z(\omega_j)$
230	367.5009	0.0000e+00	0.0000e+00
231	367.9259	1.7025e-04	0.0000e+00
232	367.9354	4.7225e-04	0.0000e+00
233	369.2067	0.0000e+00	0.0000e+00
234 *E'(z)	369.3800	0.0000e+00	2.7172e-03
235	369.4637	4.4660e-07	0.0000e+00
236	369.5608	2.1461e-08	0.0000e+00
237	372.2349	0.0000e+00	0.0000e+00
238	372.2480	0.0000e+00	1.3568e-09
239	372.4870	0.0000e+00	0.0000e+00
240	372.6904	0.0000e+00	3.0859e-06
241	372.9135	0.0000e+00	0.0000e+00
242	372.9165	0.0000e+00	9.1110e-10
243	373.6371	1.2882e-06	0.0000e+00
244	373.6807	1.1691e-05	0.0000e+00
245	374.0959	0.0000e+00	1.2271e-09
246	374.4536	1.0000e+00	0.0000e+00
247	374.5136	9.9534e-01	0.0000e+00
248	374.6015	0.0000e+00	0.0000e+00
249	375.9753	7.3998e-07	0.0000e+00
250	379.3993	0.0000e+00	0.0000e+00
251	379.4022	0.0000e+00	2.5619e-09
252	379.9541	7.2983e-09	0.0000e+00
253	379.9597	8.9618e-08	0.0000e+00
254	381.4096	8.5213e-08	0.0000e+00
255	381.4148	5.0836e-09	0.0000e+00
256	382.6173	0.0000e+00	0.0000e+00
257	382.6258	0.0000e+00	1.0278e-09
258	383.2061	1.6449e-08	0.0000e+00
259	383.2096	1.2353e-07	0.0000e+00
260	388.5925	2.9772e-09	0.0000e+00
261	388.5935	1.6798e-08	0.0000e+00
262	389.1419	0.0000e+00	0.0000e+00
263	389.1479	0.0000e+00	5.7520e-09
264	389.9698	0.0000e+00	1.1090e-09
265	390.5062	8.0234e-08	0.0000e+00
266	390.6240	2.0980e-09	0.0000e+00
267	390.7729	0.0000e+00	3.2572e-10
268	390.7914	0.0000e+00	0.0000e+00
269	393.3133	0.0000e+00	2.9506e-07
270	393.3258	0.0000e+00	0.0000e+00
271	393.9653	4.0190e-05	0.0000e+00
272	394.8126	4.3183e-10	0.0000e+00
273	394.8171	2.0629e-09	0.0000e+00
274	395.3889	2.5018e-08	0.0000e+00
275	395.4002	2.4750e-08	0.0000e+00
276	396.0900	0.0000e+00	6.9752e-08
277	396.1195	0.0000e+00	0.0000e+00
278	398.0295	0.0000e+00	0.0000e+00
279	398.0300	0.0000e+00	1.0573e-09
280	399.6199	0.0000e+00	0.0000e+00
281 *A'$_1$(z)	399.7048	0.0000e+00	2.0318e-03
282	402.2895	8.5086e-10	0.0000e+00
283	402.2957	1.5012e-08	0.0000e+00
284	402.4346	6.3738e-06	0.0000e+00
285	402.4905	1.2283e-07	0.0000e+00
286	403.9095	0.0000e+00	0.0000e+00
287	403.9191	0.0000e+00	1.4563e-07
288	404.7318	0.0000e+00	1.3542e-08
289	404.7337	0.0000e+00	0.0000e+00
290	405.9482	7.0984e-09	0.0000e+00
291	405.9501	5.7378e-10	0.0000e+00
292	407.1525	8.2134e-09	0.0000e+00
293	407.1640	6.3367e-09	0.0000e+00
294	408.8301	0.0000e+00	6.5091e-10
295	408.8305	0.0000e+00	0.0000e+00
296	411.3898	6.1117e-09	0.0000e+00
297	411.3927	6.8111e-11	0.0000e+00
298	426.8635	0.0000e+00	0.0000e+00
299	426.8718	0.0000e+00	6.3928e-08
300	441.8104	7.7818e-08	0.0000e+00
301	441.8264	2.5173e-09	0.0000e+00
302	452.8761	0.0000e+00	0.0000e+00
303	452.8877	0.0000e+00	1.3630e-04
304 *A"$_2$(x)	458.9485	5.2085e-03	0.0000e+00
305	459.1162	9.4335e-06	0.0000e+00
306	460.8882	0.0000e+00	9.9646e-01

Table S2 Frequencies (in cm^{-1}) and IR intensities (in arb. units) calculated for the $MoSe_2$ rippled supercell (continued on next page)

#	Frequency	$f_{xy}(\omega_j)$	$f_z(\omega_j)$
1	-2.6629	0.0000e+00	5.6698e-09
2	-2.5117	0.0000e+00	0.0000e+00
3	-1.9646	0.0000e+00	0.0000e+00
4	-1.1047	6.9404e-07	0.0000e+00
5	0.0000	0.0000e+00	0.0000e+00
6	0.0000	0.0000e+00	0.0000e+00
7	0.0000	0.0000e+00	0.0000e+00
8	14.6863	0.0000e+00	0.0000e+00
9	14.7134	0.0000e+00	0.0000e+00
10	16.1746	0.0000e+00	8.7662e-07
11	16.2030	0.0000e+00	0.0000e+00
12	27.7563	0.0000e+00	9.1173e-07
13	27.8300	0.0000e+00	0.0000e+00
14	35.2782	2.0900e-08	0.0000e+00
15	35.2825	0.0000e+00	0.0000e+00
16	36.1774	0.0000e+00	0.0000e+00
17	36.1873	0.0000e+00	0.0000e+00
18	51.5443	3.1899e-09	0.0000e+00
19	51.5474	0.0000e+00	0.0000e+00
20	57.6149	0.0000e+00	0.0000e+00
21	57.6172	0.0000e+00	0.0000e+00
22	61.6297	0.0000e+00	0.0000e+00
23	61.6347	0.0000e+00	0.0000e+00
24	71.8869	0.0000e+00	0.0000e+00
25	71.8898	0.0000e+00	3.8190e-09
26	76.2994	0.0000e+00	0.0000e+00
27	76.2999	0.0000e+00	0.0000e+00
28	85.0401	0.0000e+00	0.0000e+00
29	85.0459	0.0000e+00	0.0000e+00
30	89.6306	0.0000e+00	0.0000e+00
31	89.6307	0.0000e+00	0.0000e+00
32	95.2070	0.0000e+00	0.0000e+00
33	95.2091	0.0000e+00	0.0000e+00
34	102.0082	0.0000e+00	0.0000e+00
35	102.0091	0.0000e+00	0.0000e+00
36	102.1693	0.0000e+00	0.0000e+00
37	102.1704	0.0000e+00	0.0000e+00
38	107.6967	0.0000e+00	0.0000e+00
39	110.3673	0.0000e+00	0.0000e+00
40	110.6988	0.0000e+00	0.0000e+00
41	112.1514	0.0000e+00	0.0000e+00
42	112.1516	0.0000e+00	0.0000e+00
43	112.4234	0.0000e+00	0.0000e+00
44	112.4247	0.0000e+00	0.0000e+00
45	112.4459	0.0000e+00	0.0000e+00
46	112.4512	0.0000e+00	0.0000e+00
47	114.1139	0.0000e+00	0.0000e+00
48	114.1156	0.0000e+00	0.0000e+00
49	114.6704	0.0000e+00	0.0000e+00
50	114.6712	0.0000e+00	0.0000e+00
51	115.2674	0.0000e+00	0.0000e+00
52	115.2681	0.0000e+00	0.0000e+00
53	115.9309	0.0000e+00	0.0000e+00
54	115.9312	0.0000e+00	0.0000e+00
55	117.2375	0.0000e+00	0.0000e+00
56	117.2377	0.0000e+00	0.0000e+00
57	117.8539	0.0000e+00	0.0000e+00
58	117.8549	0.0000e+00	0.0000e+00
59	119.8634	0.0000e+00	0.0000e+00
60	119.8636	0.0000e+00	0.0000e+00
61	120.0330	0.0000e+00	0.0000e+00
62	120.0342	0.0000e+00	0.0000e+00
63	120.1289	0.0000e+00	0.0000e+00
64	120.1342	0.0000e+00	0.0000e+00
65	120.2579	0.0000e+00	0.0000e+00
66	120.2622	0.0000e+00	0.0000e+00
67	120.6842	0.0000e+00	0.0000e+00
68	120.6883	0.0000e+00	0.0000e+00
69	120.8324	0.0000e+00	0.0000e+00
70	120.8331	0.0000e+00	0.0000e+00
71	121.7699	0.0000e+00	0.0000e+00
72	121.7716	0.0000e+00	0.0000e+00
73	122.6488	0.0000e+00	0.0000e+00
74	122.6497	0.0000e+00	0.0000e+00
75	123.8163	0.0000e+00	0.0000e+00
76	123.8170	0.0000e+00	0.0000e+00
77	125.8856	0.0000e+00	0.0000e+00
78	125.8858	0.0000e+00	0.0000e+00
79	126.3839	0.0000e+00	0.0000e+00
80	126.3923	0.0000e+00	0.0000e+00
81	128.3717	0.0000e+00	0.0000e+00
82	128.7002	0.0000e+00	0.0000e+00
83	128.7004	0.0000e+00	0.0000e+00
84	135.4425	0.0000e+00	0.0000e+00
85	135.4438	0.0000e+00	0.0000e+00
86	136.8321	0.0000e+00	0.0000e+00
87	136.8321	0.0000e+00	0.0000e+00
88	139.5283	0.0000e+00	0.0000e+00
89	139.5284	0.0000e+00	0.0000e+00
90	141.6707	0.0000e+00	0.0000e+00
91	141.6716	0.0000e+00	0.0000e+00
92	141.9819	0.0000e+00	0.0000e+00
93	141.9840	0.0000e+00	0.0000e+00
94	142.8326	0.0000e+00	0.0000e+00
95	142.8335	0.0000e+00	0.0000e+00
96	146.1858	0.0000e+00	0.0000e+00
97	146.1861	0.0000e+00	0.0000e+00
98	149.4763	0.0000e+00	0.0000e+00
99	149.4803	0.0000e+00	0.0000e+00
100	152.3675	0.0000e+00	0.0000e+00
101	152.5578	0.0000e+00	0.0000e+00
102	153.7746	0.0000e+00	0.0000e+00
103	164.2998	2.3023e-07	0.0000e+00
104	164.3019	5.7435e-08	0.0000e+00
105	164.5174	0.0000e+00	1.1348e-07
106	164.5176	0.0000e+00	0.0000e+00
107	165.2054	8.1132e-07	0.0000e+00
108 *E"(x)	165.2447	4.0016e-05	0.0000e+00
109	165.3773	0.0000e+00	3.9538e-09
110	165.3780	0.0000e+00	0.0000e+00
111	165.5665	0.0000e+00	6.3685e-07
112	165.5727	0.0000e+00	0.0000e+00
113 *E"(y)	166.2991	1.1501e-04	0.0000e+00
114	166.3314	1.1228e-06	0.0000e+00
115	168.0348	0.0000e+00	0.0000e+00
116	168.0371	0.0000e+00	0.0000e+00
117	168.8408	0.0000e+00	2.3263e-09
118	168.8453	0.0000e+00	0.0000e+00
119	172.0236	0.0000e+00	0.0000e+00
120	172.0241	0.0000e+00	0.0000e+00
121	173.5651	2.7199e-08	0.0000e+00
122	173.5659	0.0000e+00	0.0000e+00
123	176.5753	0.0000e+00	0.0000e+00
124	176.5777	0.0000e+00	0.0000e+00
125	179.5178	0.0000e+00	0.0000e+00
126	180.2997	0.0000e+00	0.0000e+00
127	180.3003	0.0000e+00	0.0000e+00
128	180.8233	0.0000e+00	0.0000e+00
129	180.8240	0.0000e+00	0.0000e+00
130	182.2798	0.0000e+00	0.0000e+00
131	182.6072	0.0000e+00	0.0000e+00
132	184.1532	0.0000e+00	0.0000e+00
133	184.1554	0.0000e+00	0.0000e+00
134	186.1518	0.0000e+00	0.0000e+00
135	186.1523	0.0000e+00	0.0000e+00
136	186.5818	0.0000e+00	0.0000e+00
137	186.5829	0.0000e+00	0.0000e+00
138	186.7730	0.0000e+00	0.0000e+00
139	186.7735	0.0000e+00	0.0000e+00
140	187.1726	0.0000e+00	0.0000e+00
141	187.1764	0.0000e+00	0.0000e+00
142	187.7026	0.0000e+00	0.0000e+00
143	187.7034	0.0000e+00	0.0000e+00
144	188.2704	0.0000e+00	0.0000e+00
145	188.2707	0.0000e+00	0.0000e+00
146	188.4532	0.0000e+00	0.0000e+00
147	188.4541	0.0000e+00	0.0000e+00
148	192.2173	0.0000e+00	0.0000e+00
149	192.2174	0.0000e+00	0.0000e+00
150	192.4596	0.0000e+00	0.0000e+00
151	192.4606	0.0000e+00	0.0000e+00
152	194.5190	0.0000e+00	0.0000e+00

#	Frequency	$f_{xy}(\omega_j)$	$f_z(\omega_j)$
153	194.5268	0.0000e+00	0.0000e+00
154	194.5273	0.0000e+00	0.0000e+00
155	194.5752	0.0000e+00	0.0000e+00
156	194.5758	0.0000e+00	0.0000e+00
157	194.5888	0.0000e+00	0.0000e+00
158	194.6389	0.0000e+00	0.0000e+00
159	194.9594	0.0000e+00	0.0000e+00
160	194.9616	0.0000e+00	0.0000e+00
161	194.9821	0.0000e+00	0.0000e+00
162	194.9920	0.0000e+00	0.0000e+00
163	195.0167	0.0000e+00	0.0000e+00
164	195.0168	0.0000e+00	0.0000e+00
165	196.1237	0.0000e+00	0.0000e+00
166	196.1242	0.0000e+00	0.0000e+00
167	196.1625	0.0000e+00	0.0000e+00
168	196.1636	0.0000e+00	0.0000e+00
169	196.3572	0.0000e+00	0.0000e+00
170	196.3587	0.0000e+00	0.0000e+00
171	196.8589	0.0000e+00	0.0000e+00
172	196.8613	0.0000e+00	0.0000e+00
173	198.2623	0.0000e+00	0.0000e+00
174	198.2641	0.0000e+00	0.0000e+00
175	200.8625	0.0000e+00	0.0000e+00
176	200.8627	0.0000e+00	0.0000e+00
177	201.9206	0.0000e+00	0.0000e+00
178	201.9208	0.0000e+00	0.0000e+00
179	202.4013	0.0000e+00	0.0000e+00
180	202.4016	0.0000e+00	0.0000e+00
181	202.6970	0.0000e+00	0.0000e+00
182	202.6971	0.0000e+00	0.0000e+00
183	203.5375	0.0000e+00	0.0000e+00
184	203.5389	0.0000e+00	0.0000e+00
185	206.3360	0.0000e+00	0.0000e+00
186	206.3453	0.0000e+00	0.0000e+00
187	207.7706	0.0000e+00	0.0000e+00
188	211.8350	0.0000e+00	0.0000e+00
189	211.8353	0.0000e+00	0.0000e+00
190	217.1501	0.0000e+00	0.0000e+00
191	217.1509	0.0000e+00	0.0000e+00
192	218.5335	0.0000e+00	0.0000e+00
193	218.5344	0.0000e+00	0.0000e+00
194	219.0516	0.0000e+00	0.0000e+00
195	219.0535	0.0000e+00	0.0000e+00
196	219.9614	0.0000e+00	0.0000e+00
197	219.9623	0.0000e+00	0.0000e+00
198	222.2880	0.0000e+00	4.8598e-09
199	222.2904	0.0000e+00	0.0000e+00
200	226.1703	8.3005e-06	0.0000e+00
201	226.1705	4.9376e-06	0.0000e+00
202	230.6795	0.0000e+00	0.0000e+00
203 *A'$_1$(z)	230.7519	0.0000e+00	1.2680e-05
204	233.4271	1.8821e-05	0.0000e+00
205	249.4671	0.0000e+00	0.0000e+00
206	249.4673	0.0000e+00	0.0000e+00
207	250.5295	1.2693e-09	0.0000e+00
208	250.5298	0.0000e+00	0.0000e+00
209	255.3279	1.2084e-09	0.0000e+00
210	255.3289	0.0000e+00	0.0000e+00
211	256.9785	0.0000e+00	0.0000e+00
212	256.9785	0.0000e+00	0.0000e+00
213	264.6738	0.0000e+00	0.0000e+00
214	264.6739	0.0000e+00	0.0000e+00
215	265.3222	0.0000e+00	0.0000e+00
216	265.3223	0.0000e+00	0.0000e+00
217	267.9888	0.0000e+00	0.0000e+00
218	267.9892	0.0000e+00	0.0000e+00
219	269.2317	0.0000e+00	0.0000e+00
220	269.8267	0.0000e+00	0.0000e+00
221	269.8267	0.0000e+00	0.0000e+00
222	269.8656	0.0000e+00	0.0000e+00
223	269.8658	0.0000e+00	0.0000e+00
224	270.1820	0.0000e+00	0.0000e+00
225	270.1820	0.0000e+00	0.0000e+00
226	270.5389	1.2482e-09	0.0000e+00
227	270.5956	0.0000e+00	0.0000e+00
228	270.9801	0.0000e+00	0.0000e+00
229	270.9818	0.0000e+00	0.0000e+00
230	277.2789	5.7531e-03	0.0000e+00
231	277.2791	6.0488e-03	0.0000e+00
232	277.4235	0.0000e+00	0.0000e+00
233	277.4882	0.0000e+00	1.3966e-04
234	277.7334	9.9393e-01	0.0000e+00
235	277.8259	9.9558e-01	0.0000e+00
236	278.8554	0.0000e+00	9.8602e-09
237	278.8563	0.0000e+00	0.0000e+00
238	279.2194	2.3366e-07	0.0000e+00
239	279.2202	1.2872e-07	0.0000e+00
240	281.2949	0.0000e+00	0.0000e+00
241	281.2958	0.0000e+00	0.0000e+00
242	282.0133	3.1367e-09	0.0000e+00
243	282.0137	0.0000e+00	0.0000e+00
244	283.5714	0.0000e+00	0.0000e+00
245	283.6668	5.5553e-08	0.0000e+00
246	284.0780	0.0000e+00	0.0000e+00
247	284.0787	0.0000e+00	0.0000e+00
248	284.3158	0.0000e+00	0.0000e+00
249	284.3165	0.0000e+00	0.0000e+00
250	284.7569	0.0000e+00	0.0000e+00
251	284.7606	0.0000e+00	0.0000e+00
252	285.0748	0.0000e+00	0.0000e+00
253	285.6463	1.8453e-08	0.0000e+00
254	285.6464	0.0000e+00	0.0000e+00
255	285.7199	0.0000e+00	0.0000e+00
256	285.7221	0.0000e+00	0.0000e+00
257	285.9973	0.0000e+00	0.0000e+00
258	285.9975	4.0972e-09	0.0000e+00
259	286.3782	0.0000e+00	2.4080e-09
260	286.3802	0.0000e+00	0.0000e+00
261 *E'(z)	286.3858	0.0000e+00	1.1638e-03
262	286.4566	0.0000e+00	0.0000e+00
263	286.5439	3.9181e-09	0.0000e+00
264	286.5444	0.0000e+00	0.0000e+00
265	286.9856	2.6576e-08	0.0000e+00
266	286.9861	0.0000e+00	0.0000e+00
267	287.3942	0.0000e+00	0.0000e+00
268	287.3954	0.0000e+00	0.0000e+00
269	288.0934	0.0000e+00	0.0000e+00
270	288.0967	0.0000e+00	0.0000e+00
271	288.9074	0.0000e+00	0.0000e+00
272	288.9085	0.0000e+00	0.0000e+00
273	289.3836	0.0000e+00	0.0000e+00
274	289.3837	2.4984e-09	0.0000e+00
275	289.6797	0.0000e+00	0.0000e+00
276	289.7007	0.0000e+00	0.0000e+00
277	290.7716	0.0000e+00	0.0000e+00
278	290.7867	3.1457e-09	0.0000e+00
279	290.7873	0.0000e+00	0.0000e+00
280	291.0347	0.0000e+00	0.0000e+00
281	291.3793	0.0000e+00	0.0000e+00
282	291.5225	0.0000e+00	0.0000e+00
283	291.5239	0.0000e+00	0.0000e+00
284	292.0997	0.0000e+00	0.0000e+00
285	292.1013	0.0000e+00	0.0000e+00
286	294.8619	0.0000e+00	0.0000e+00
287	294.8656	0.0000e+00	0.0000e+00
288	296.6353	0.0000e+00	0.0000e+00
289	296.6354	0.0000e+00	1.0003e-08
290	298.1685	1.9924e-06	0.0000e+00
291	298.1707	2.6434e-06	0.0000e+00
292	300.0483	0.0000e+00	0.0000e+00
293	300.0488	0.0000e+00	0.0000e+00
294	305.3310	0.0000e+00	0.0000e+00
295	305.3354	0.0000e+00	0.0000e+00
296	310.4386	0.0000e+00	0.0000e+00
297	310.4386	0.0000e+00	0.0000e+00
298	315.8700	0.0000e+00	1.4546e-09
299	315.8747	0.0000e+00	0.0000e+00
300	322.6883	6.4744e-09	0.0000e+00
301	322.6889	5.3295e-09	0.0000e+00
302	331.0249	0.0000e+00	1.9422e-04
303	331.0312	0.0000e+00	0.0000e+00
304 *A"$_2$(x)	338.4135	1.4042e-03	0.0000e+00
305	338.6413	1.1579e-04	0.0000e+00
306	341.6002	0.0000e+00	1.0000e+00