Supplementary Material for "Physics-in-the-loop Bayesian optimization enables autonomous low-energy electron diffraction-I(V) analysis"

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Table S1: Parameters of DFT calculations

Parameters	**Ag(100)-(1×1) surface**	**$Fe_2O_3(1\bar{1}02)$-(1×1)**
K-mesh	$10 \times 10 \times 4$	$7 \times 7 \times 1$
Energy convergence criterion	10^{-6} eV	10^{-6} eV
Cutoff energy	500 eV	500 eV
LDAUTYPE	–	Dudarev's approach [1]
LDAUU	–	4
LDAUJ	–	0

Table S2: Hyperparameters and optimization parameters in physics-in-the-loop Bayesian optimization

Hyperparameters	**Ag(100)-(1×1) surface**	**$Fe_2O_3(1\bar{1}02)$-(1×1)**
Number of optimized parameters	13	53
Surrogate model	SingleTaskGP (Marten 2.5)	SingleTaskGP (Marten 2.5)
Acquisition Function	qUCB (β=1)	Thompson sampling
Number of trust regions	1	4
Optimized parameters		
Number of optimized coordinates	12	50
Length of coordinates interval	0.3 Å	0.3 Å
Length of VIBROCC interval	0.05Å (Ag)	0.05 Å (Fe and O)
Incidence angle	-	0° - 2°

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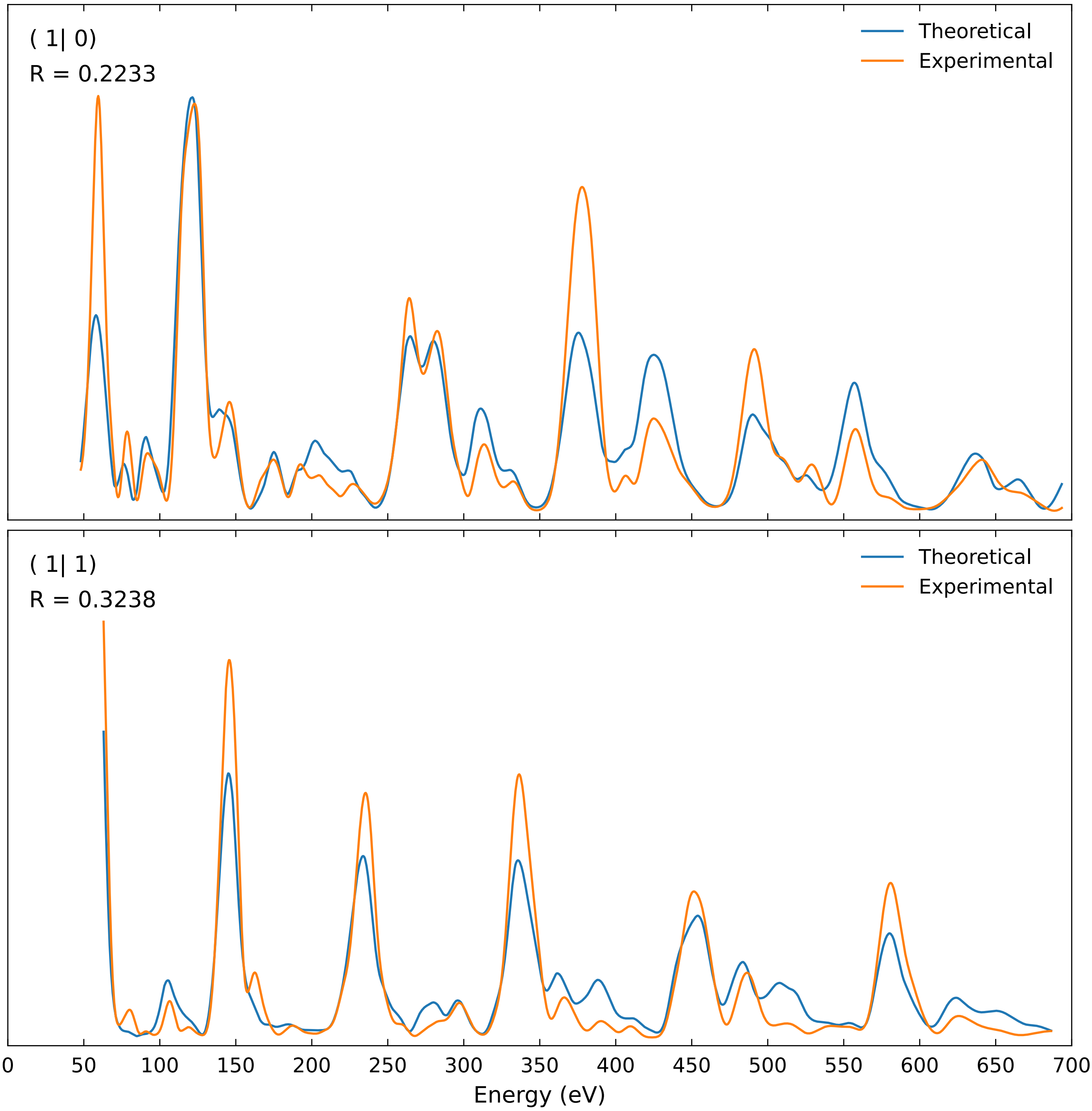
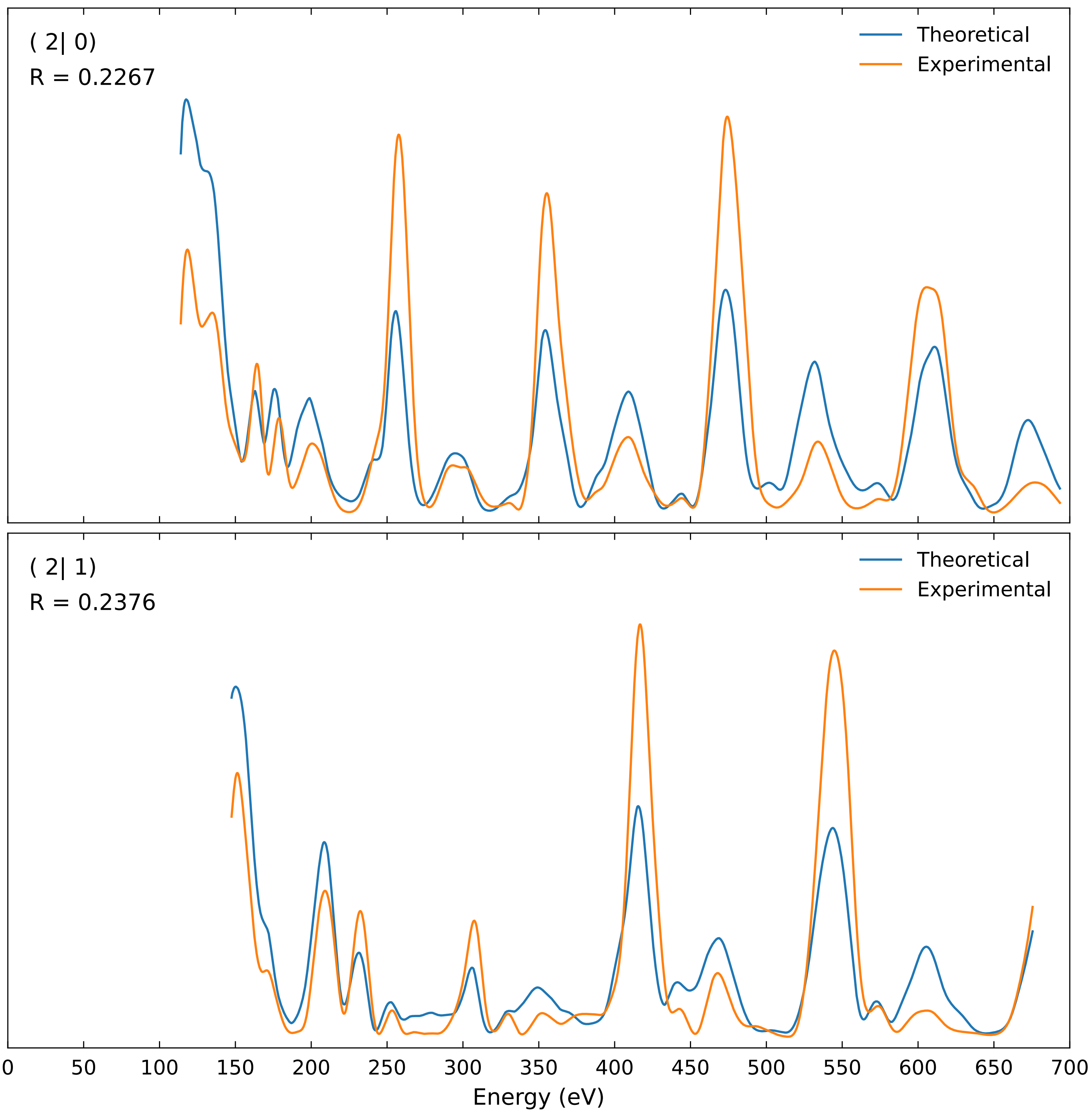
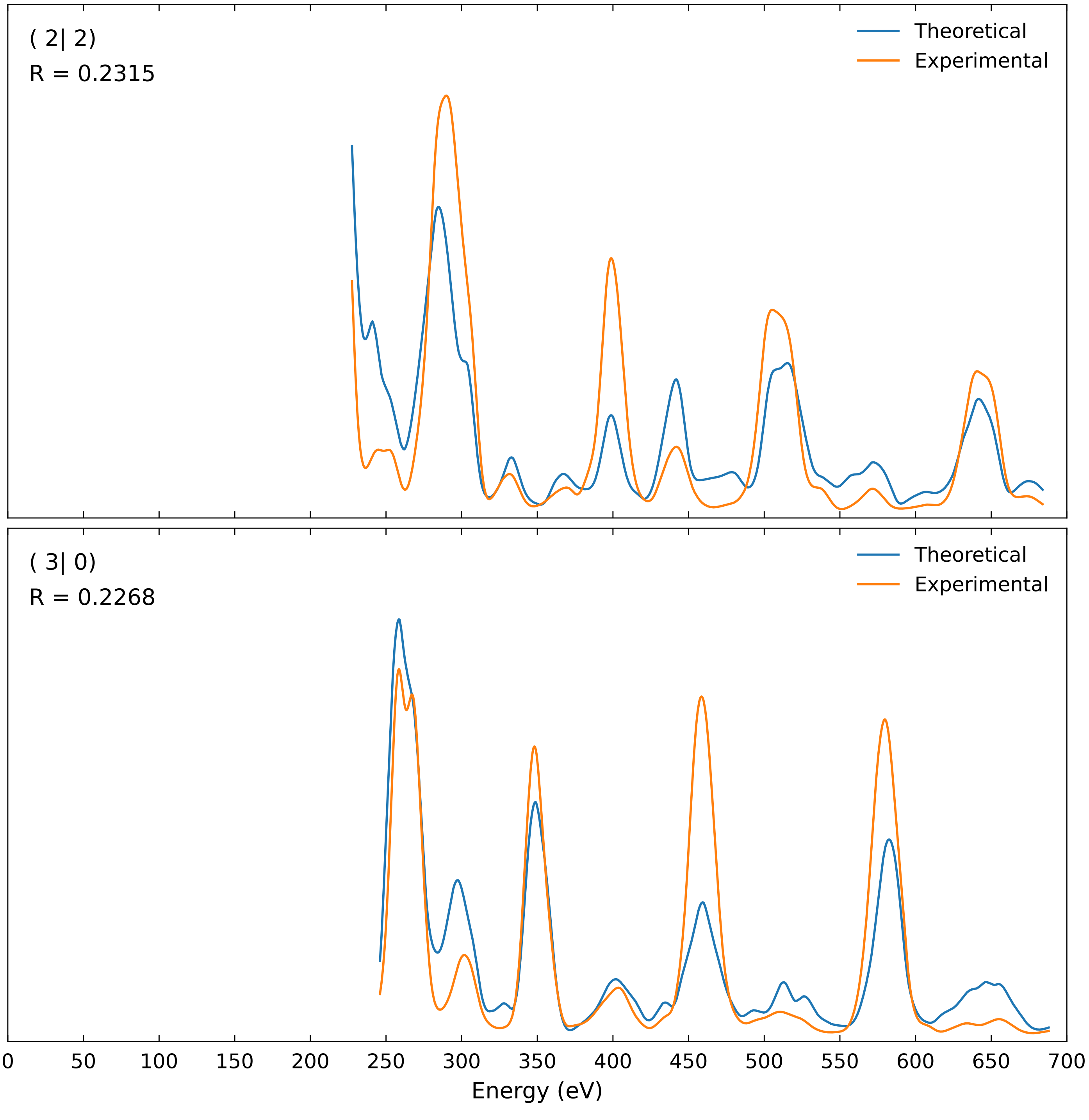
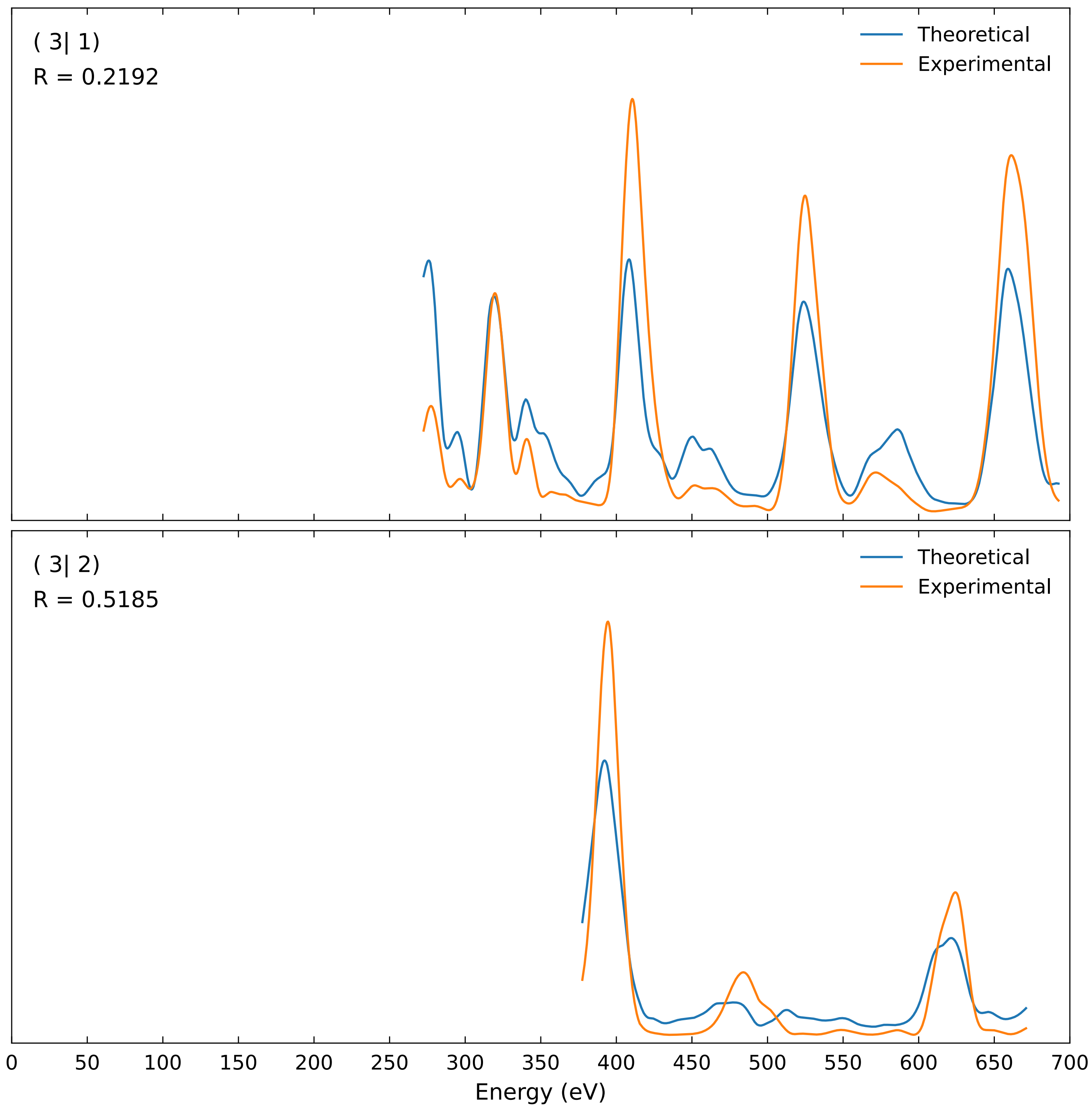
Figure S1: Matching results of LEED-I(V) curves for the Ag(100)-(1×1) surface at stage A in Fig. 2a. Experimental and calculated LEED-I(V) curves are shown for all diffraction beams.

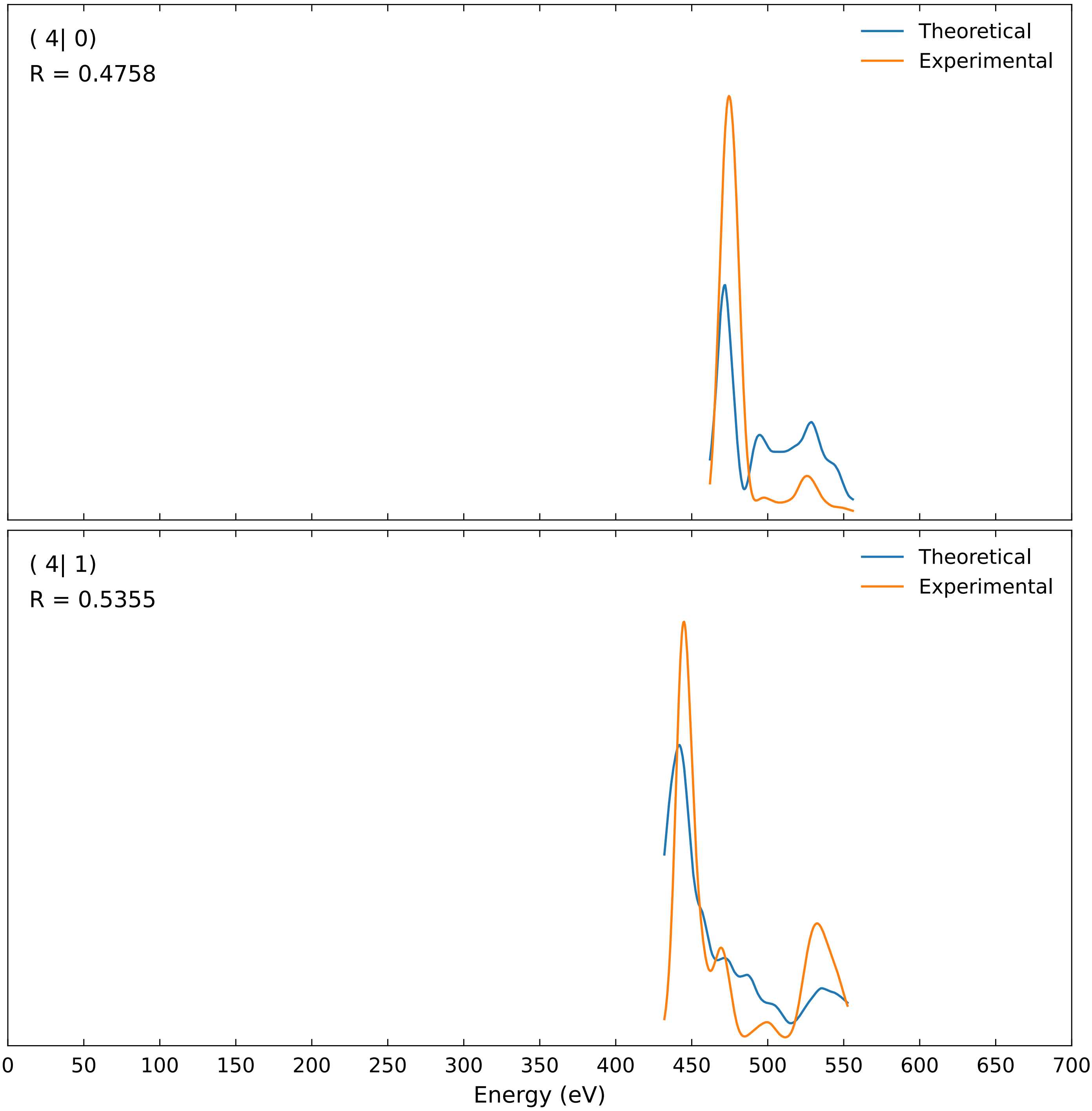




(2| 0)
R = 0.2267
Theoretical
Experimental
(2| 1)
R = 0.2376
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

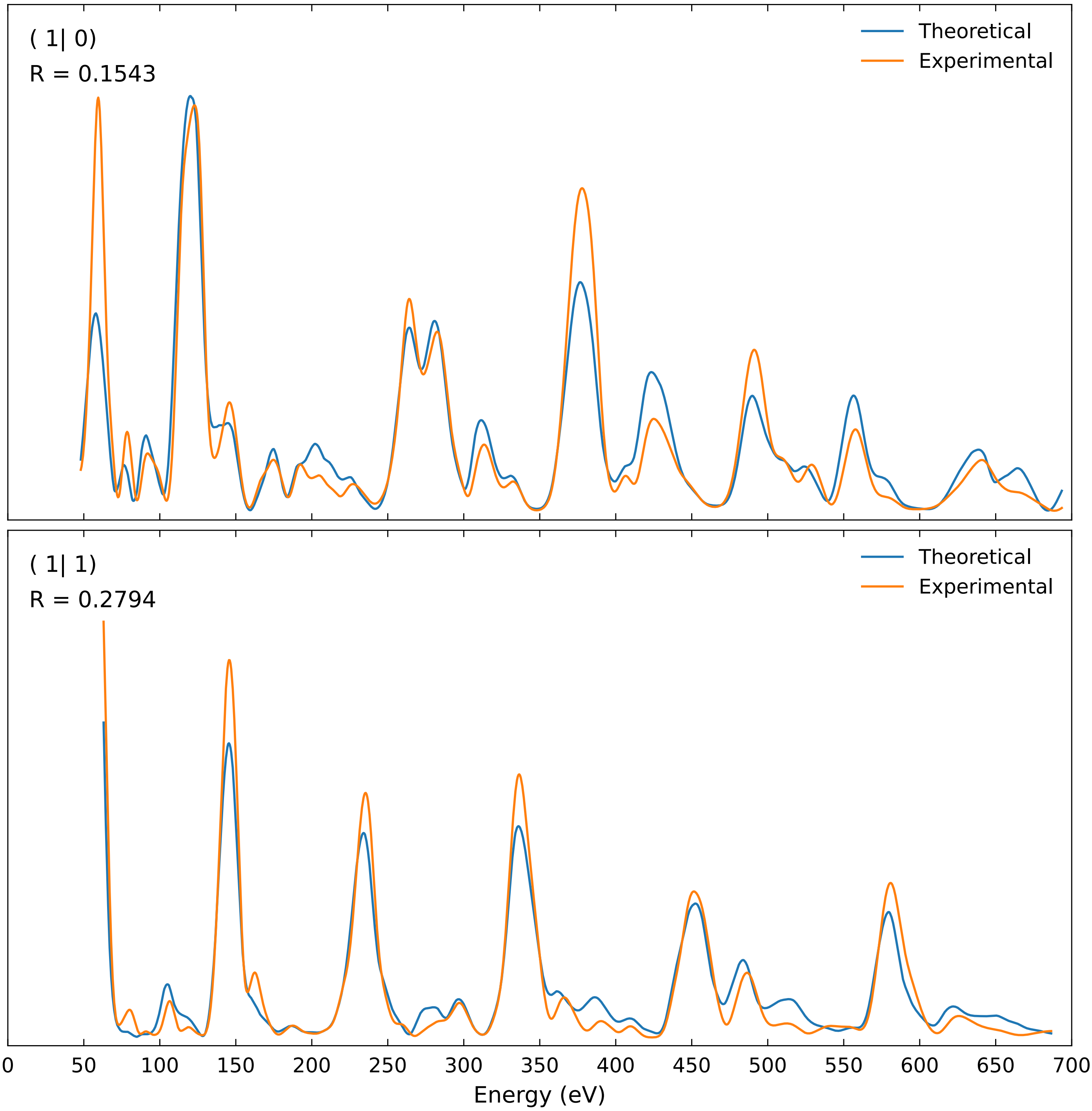

(2| 2)
R = 0.2315
Theoretical
Experimental
(3| 0)
R = 0.2268
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

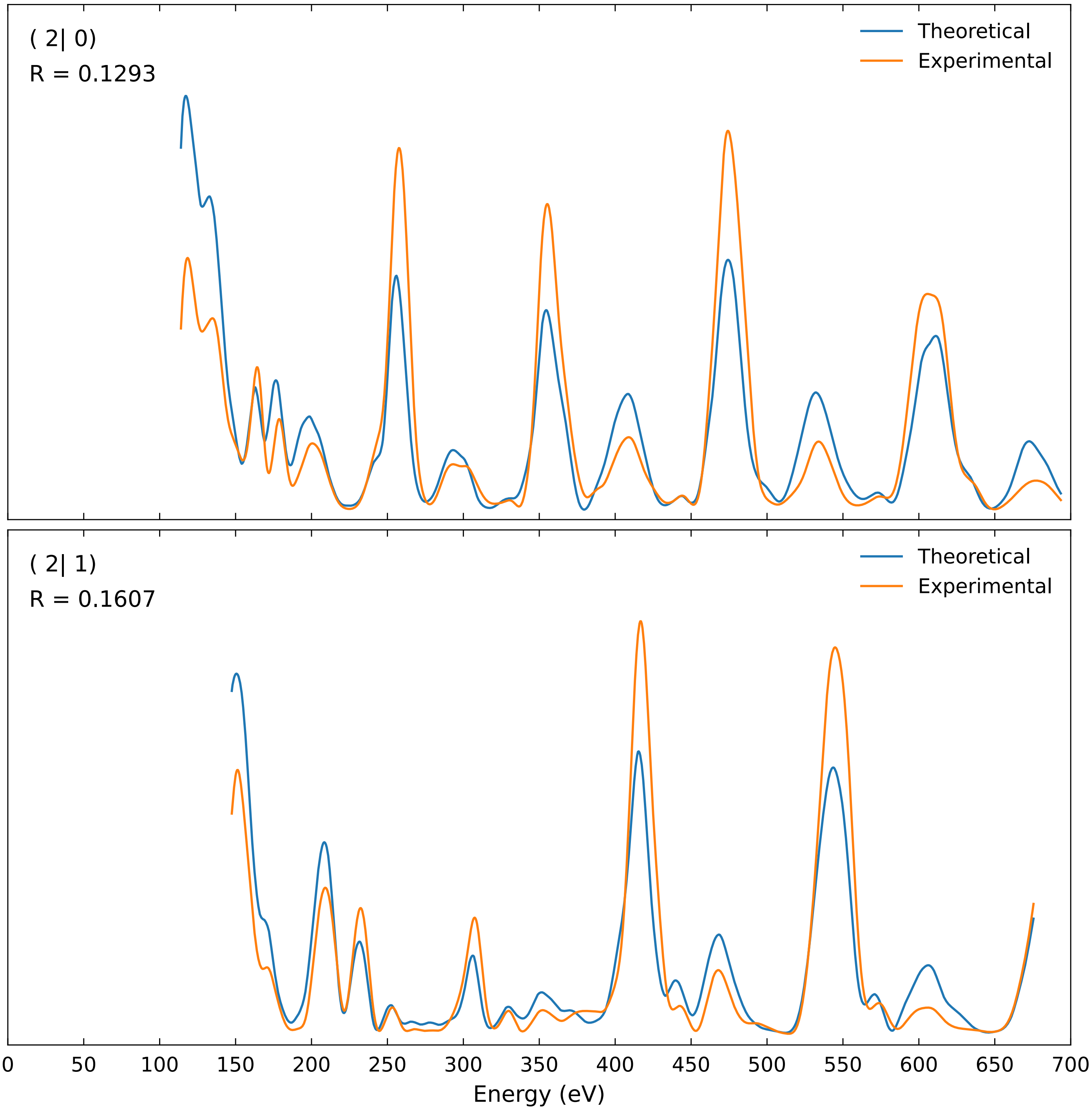

(3| 1)
R = 0.2192
Theoretical
Experimental
(3| 2)
R = 0.5185
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)



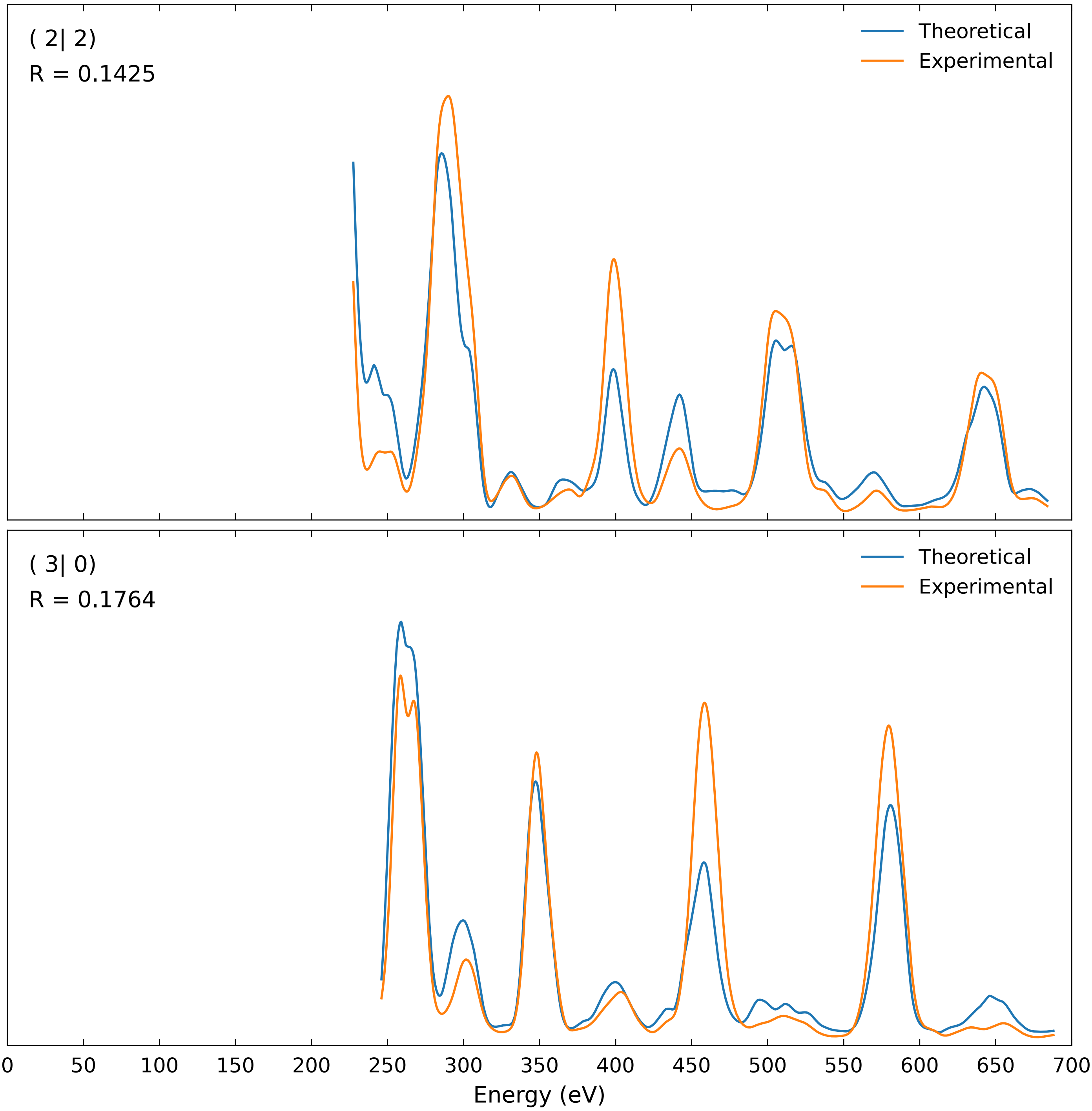
(4| 0)
R = 0.4758
Theoretical
Experimental
(4| 1)
R = 0.5355
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

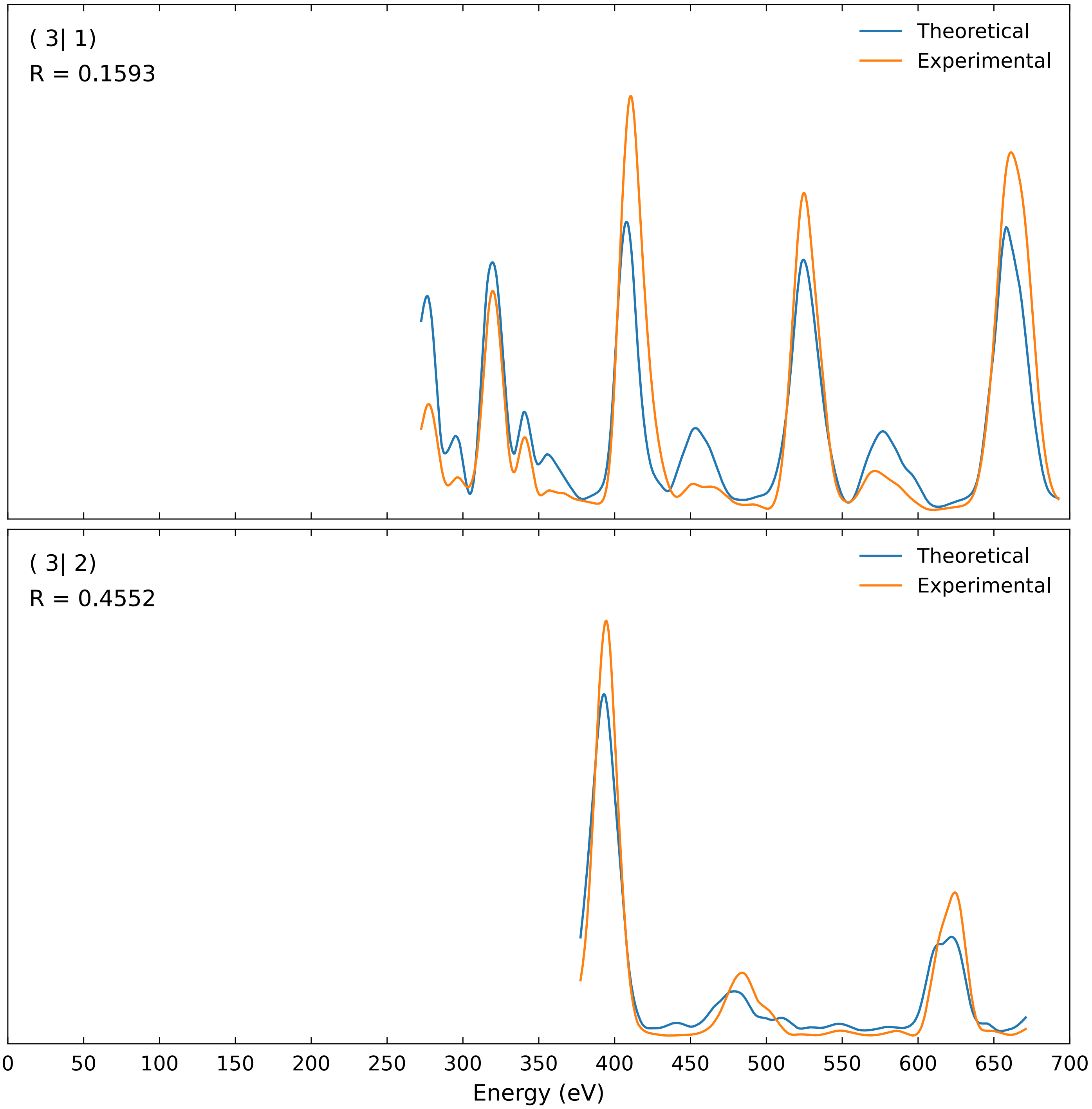
Figure S2: Matching results of LEED-I(V) curves for the Ag(100)-(1×1) surface at stage B in Fig. 2a. Experimental and calculated LEED-I(V) curves are shown for all diffraction beams.



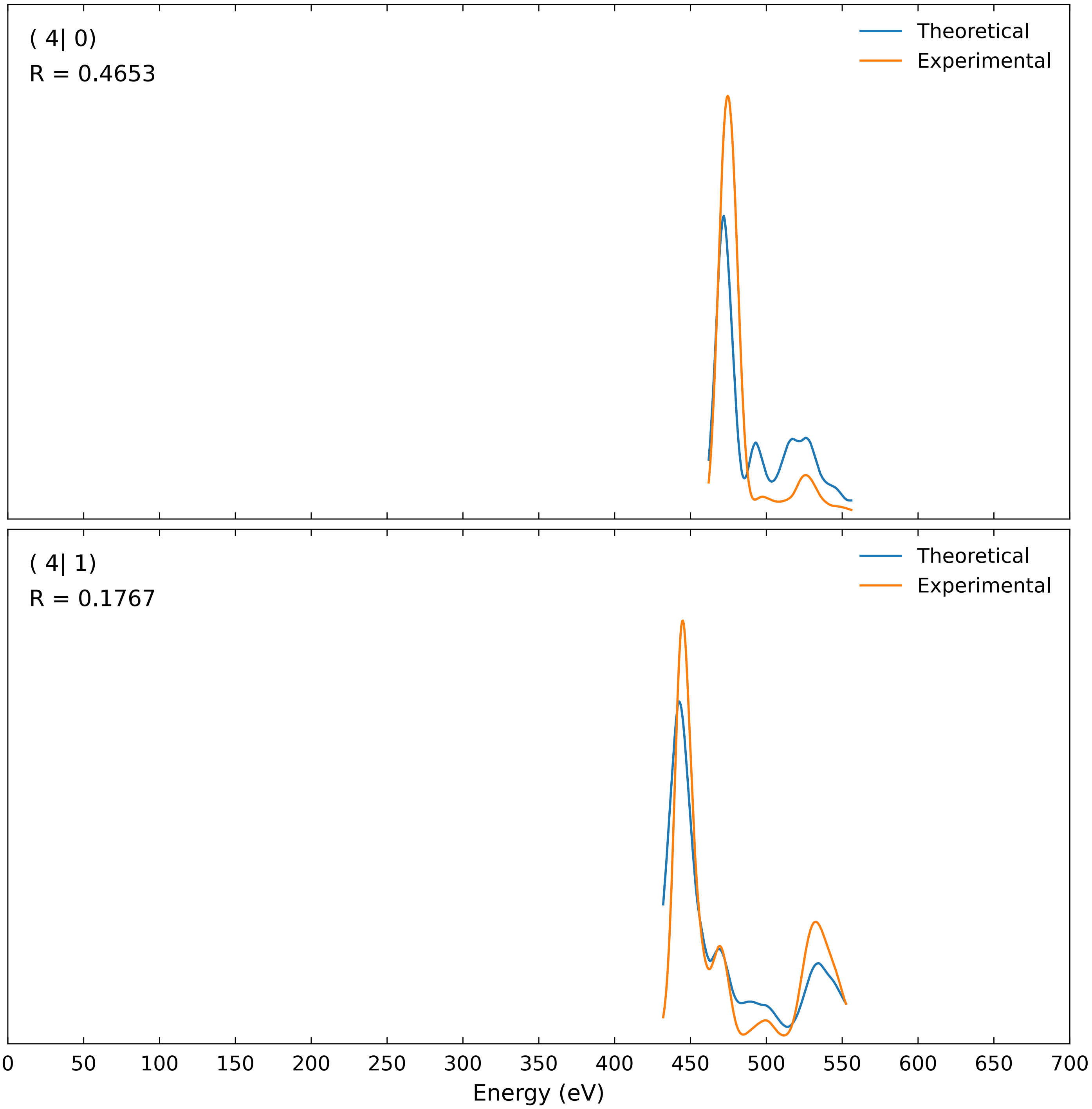


(2| 0)
R = 0.1293
Theoretical
Experimental
(2| 1)
R = 0.1607
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)


(2| 2)
R = 0.1425
Theoretical
Experimental
(3| 0)
R = 0.1764
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

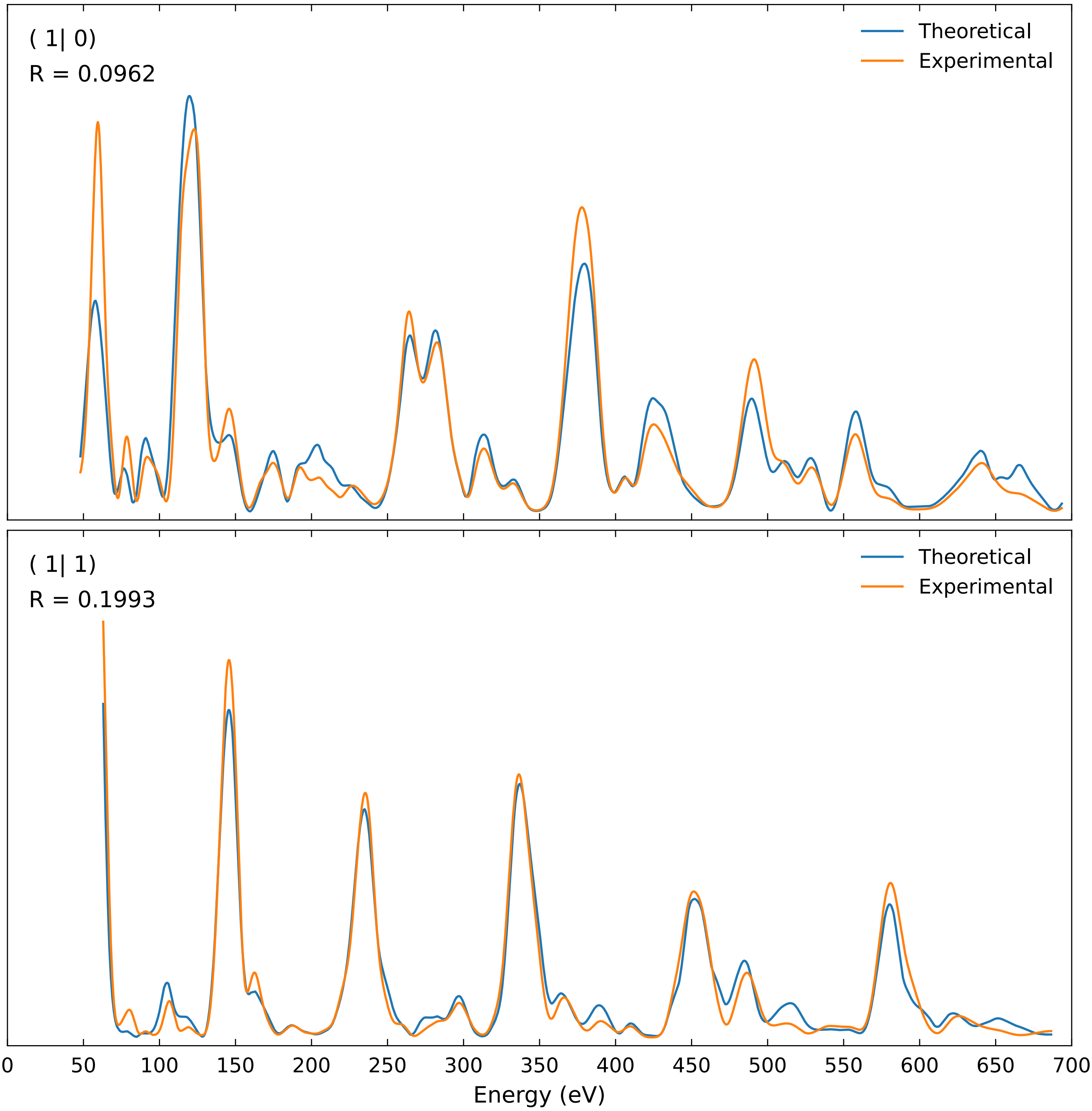


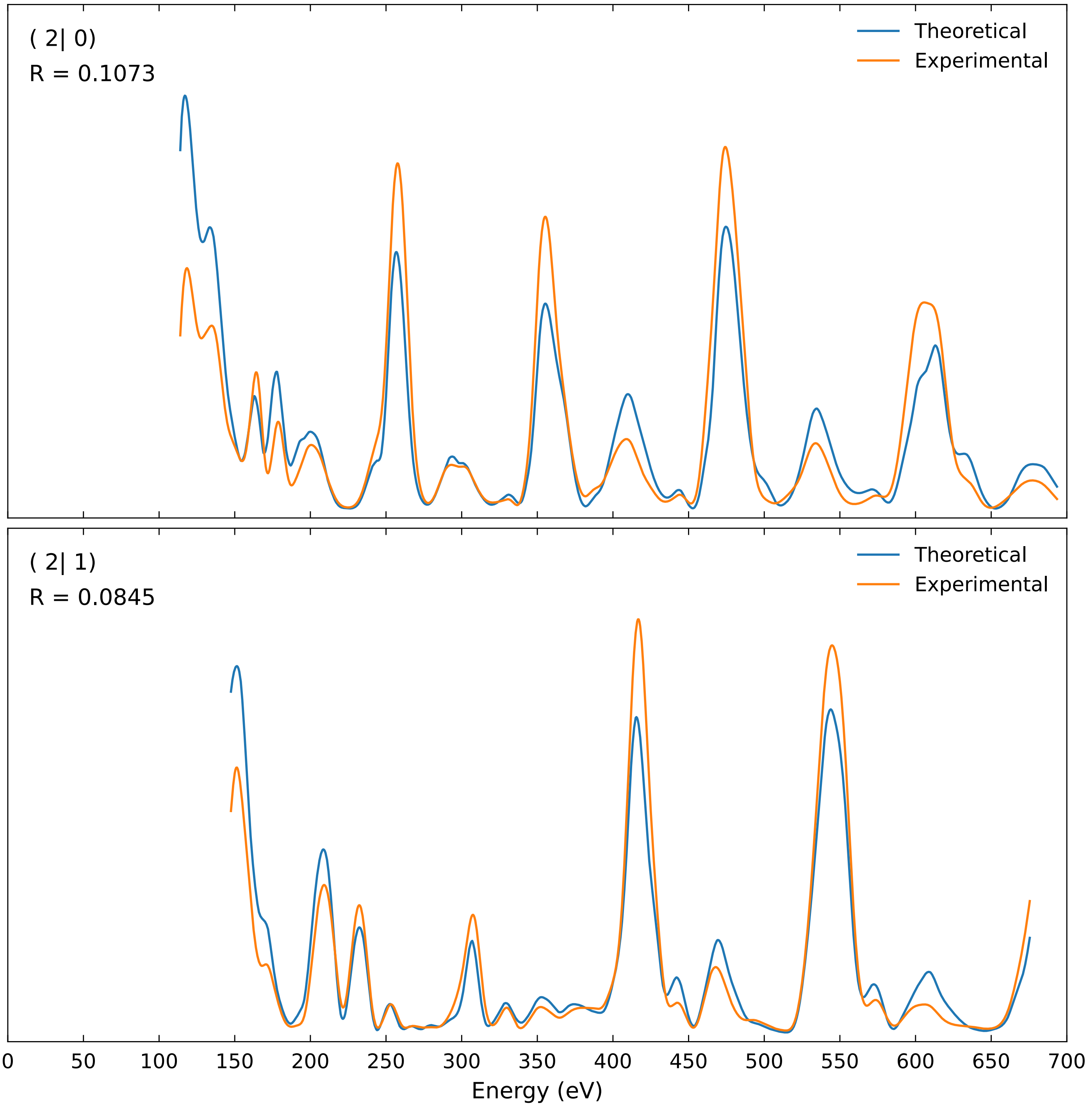
(3| 1)
R = 0.1593
Theoretical
Experimental
(3| 2)
R = 0.4552
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

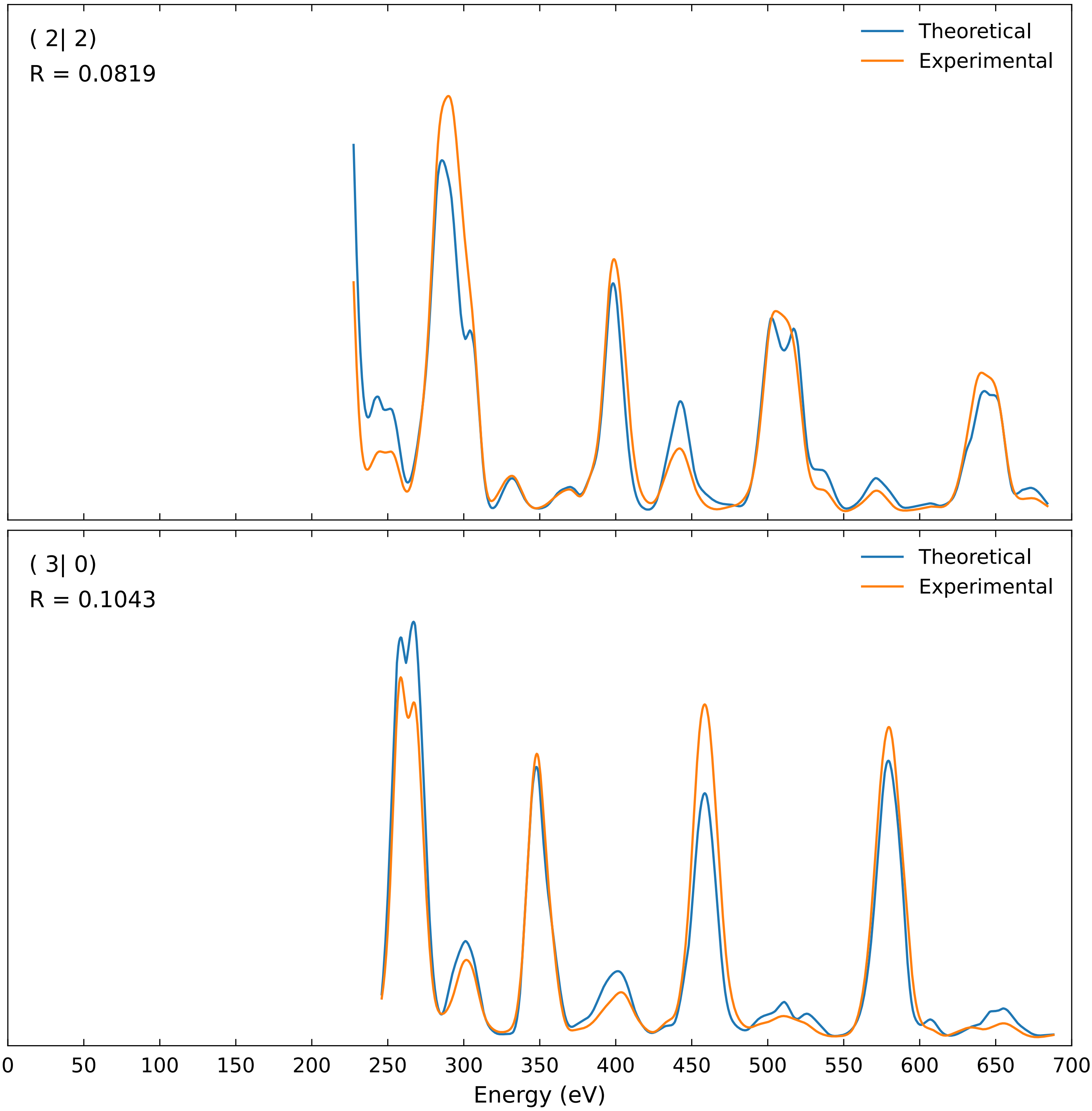


(4| 0)
R = 0.4653
Theoretical
Experimental
(4| 1)
R = 0.1767
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

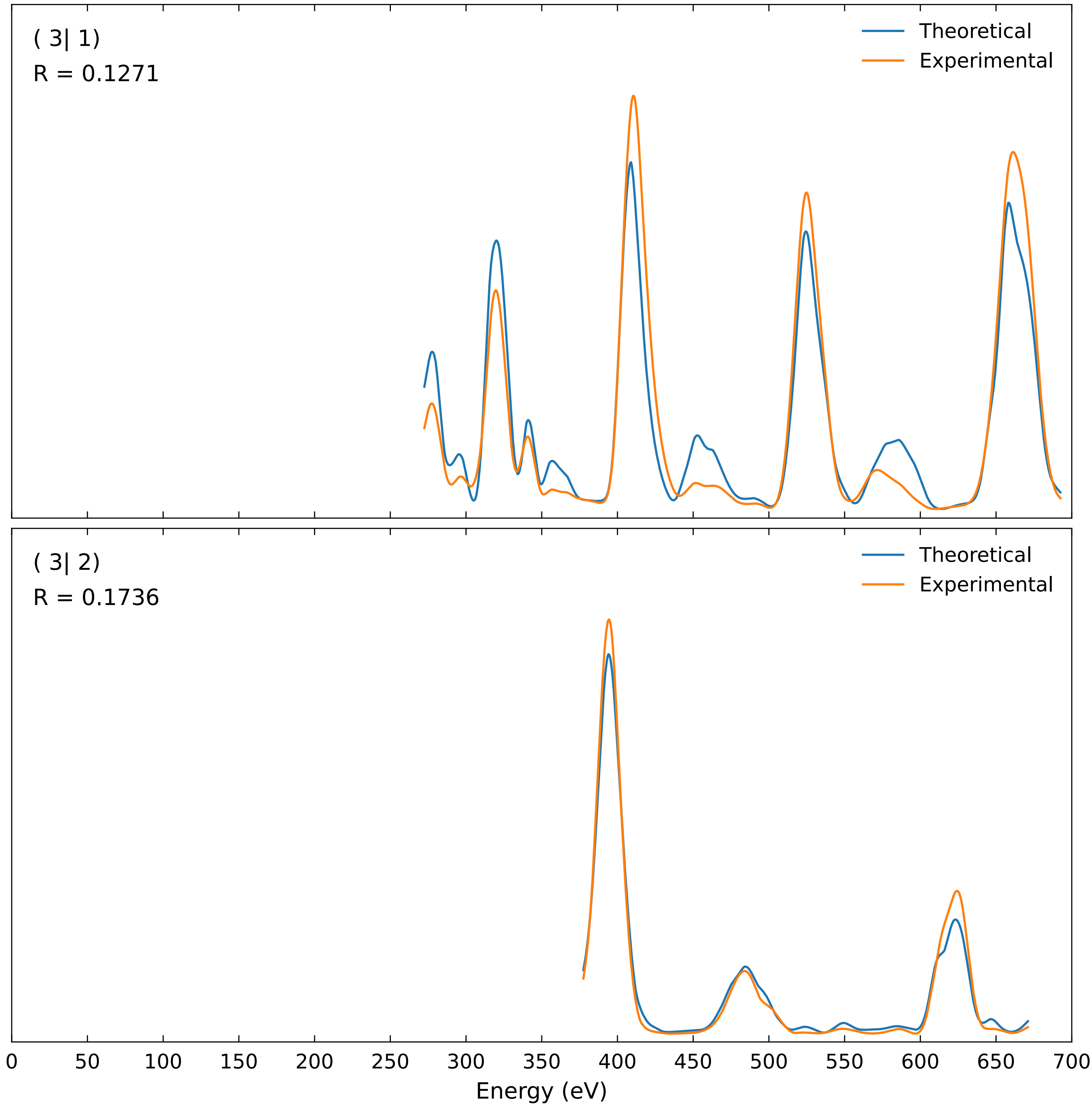
Figure S3: Matching results of LEED-I(V) curves for the Ag(100)-(1×1) surface at stage B in Fig. 2a. Experimental and calculated LEED-I(V) curves are shown for all diffraction beams.



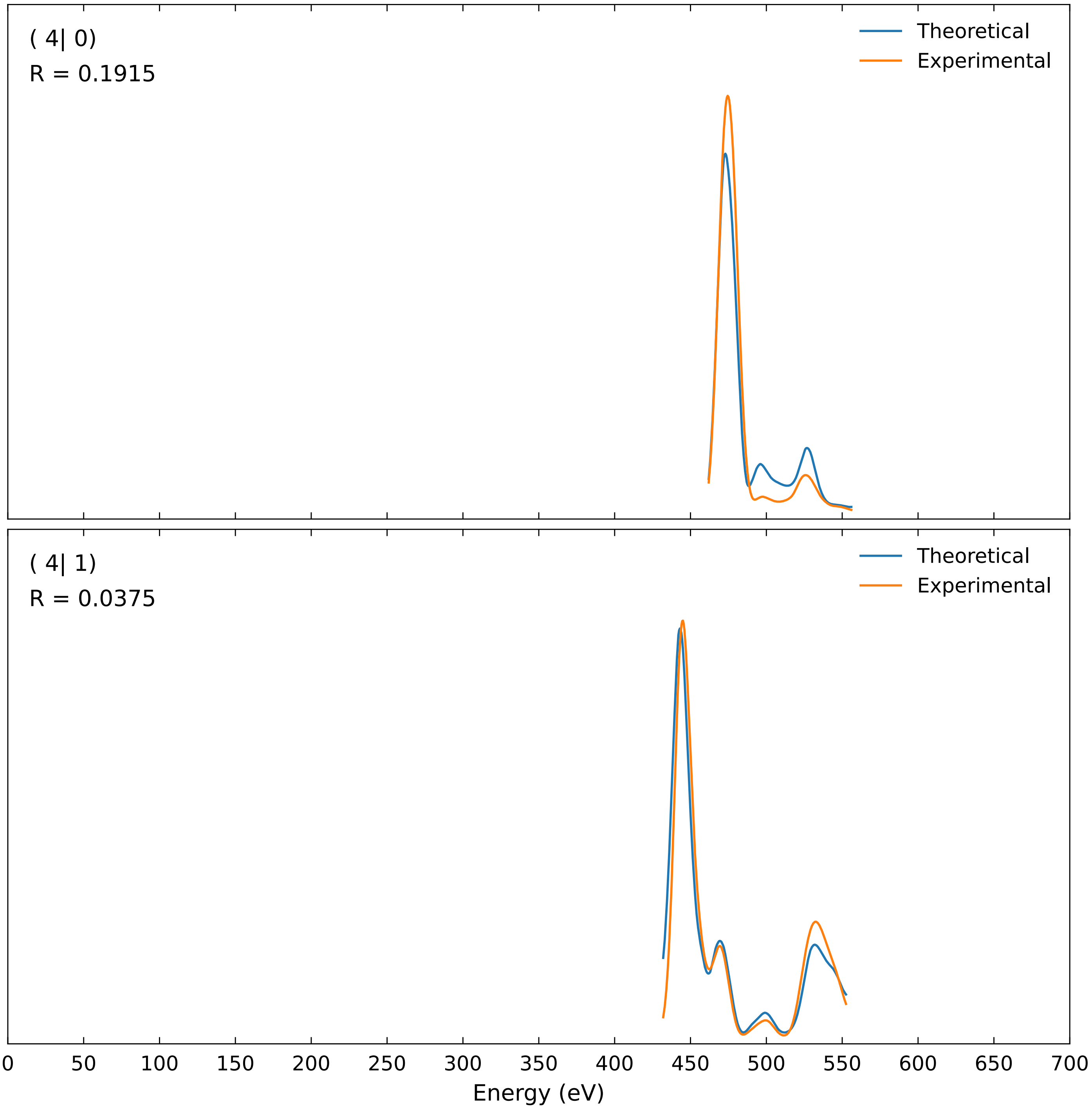

(2| 0)
R = 0.1073
Theoretical
Experimental
(2| 1)
R = 0.0845
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)



(2| 2)
R = 0.0819
Theoretical
Experimental
(3| 0)
R = 0.1043
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

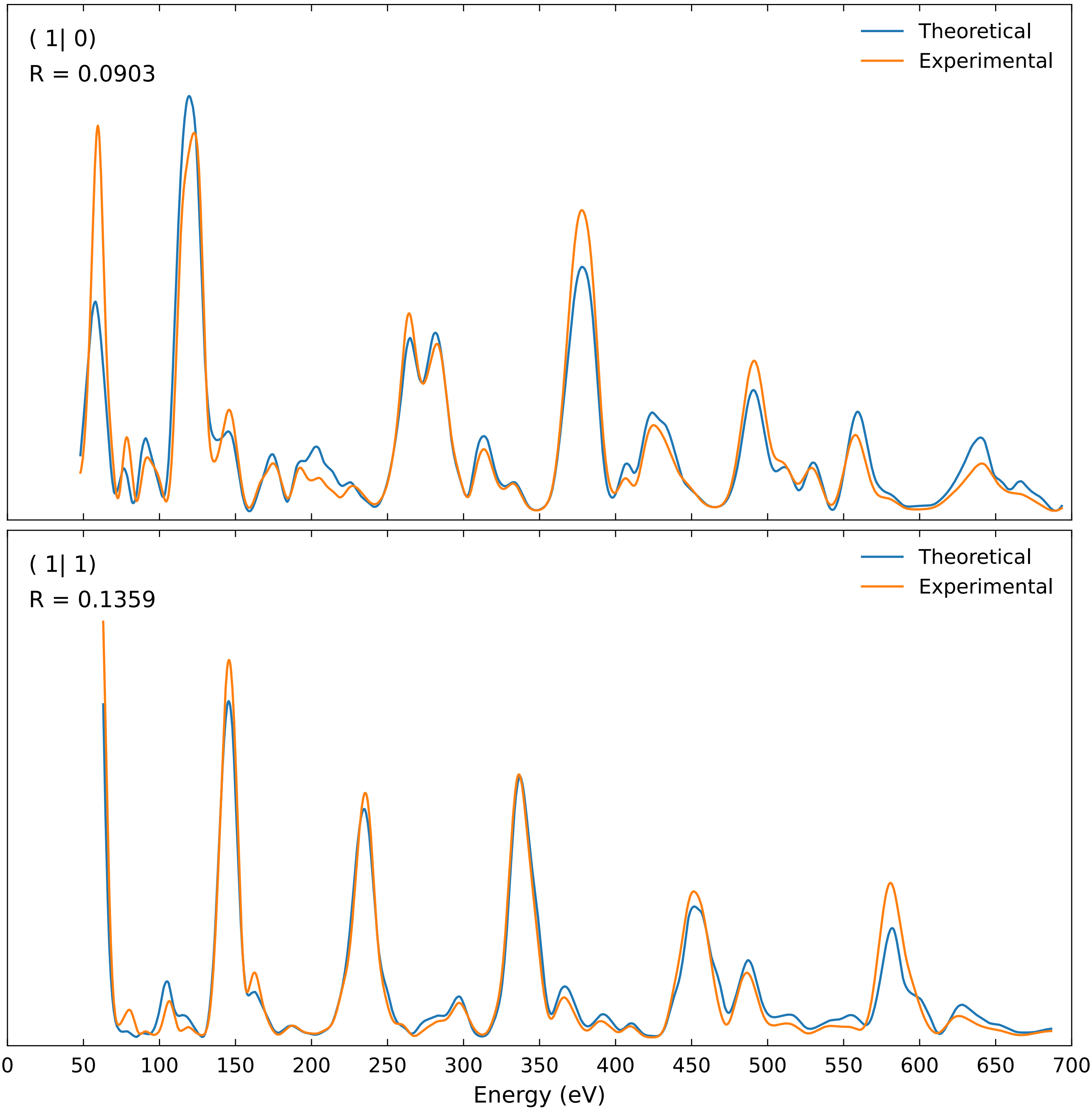


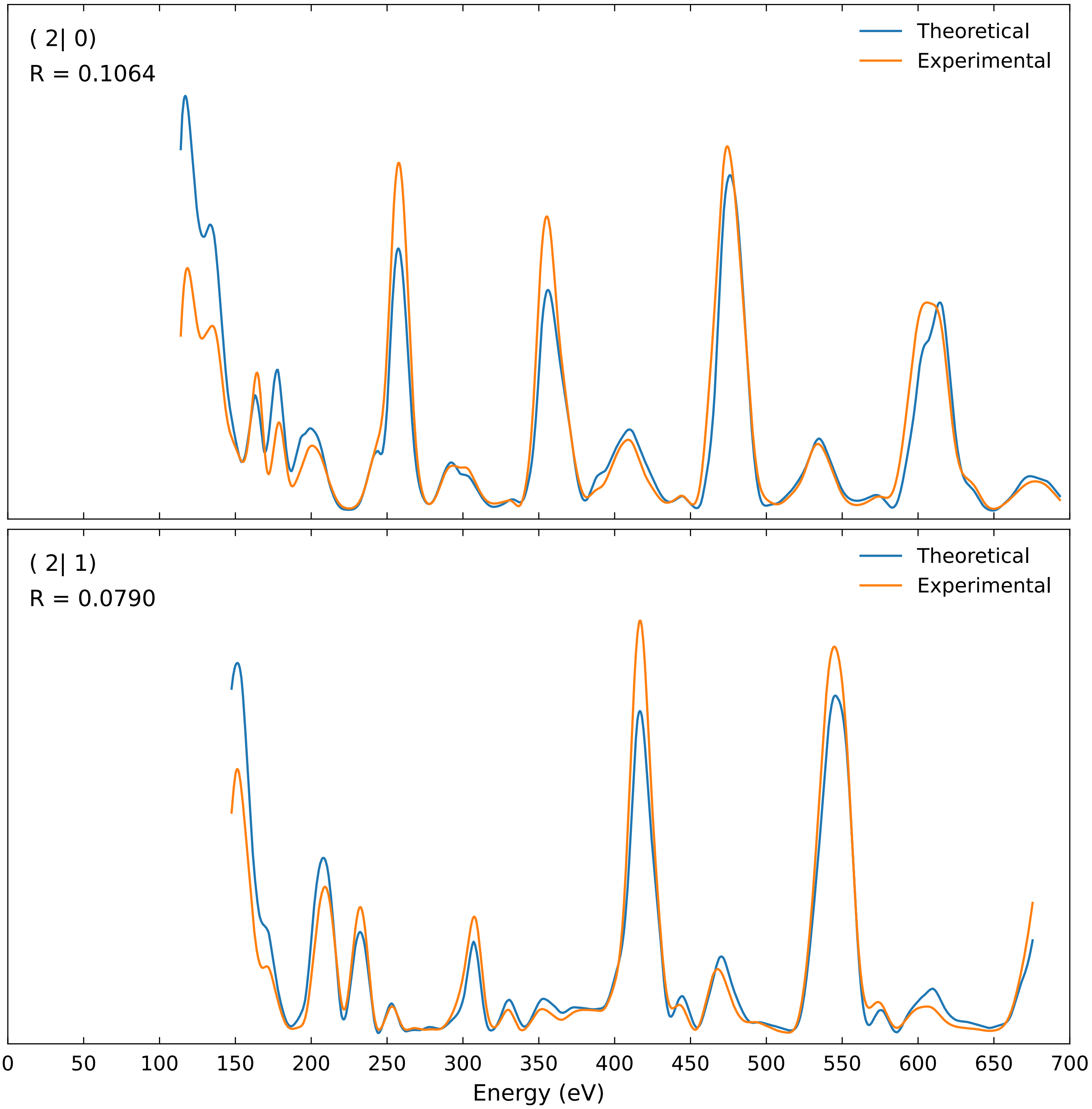
(3| 1)
R = 0.1271
Theoretical
Experimental
(3| 2)
R = 0.1736
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

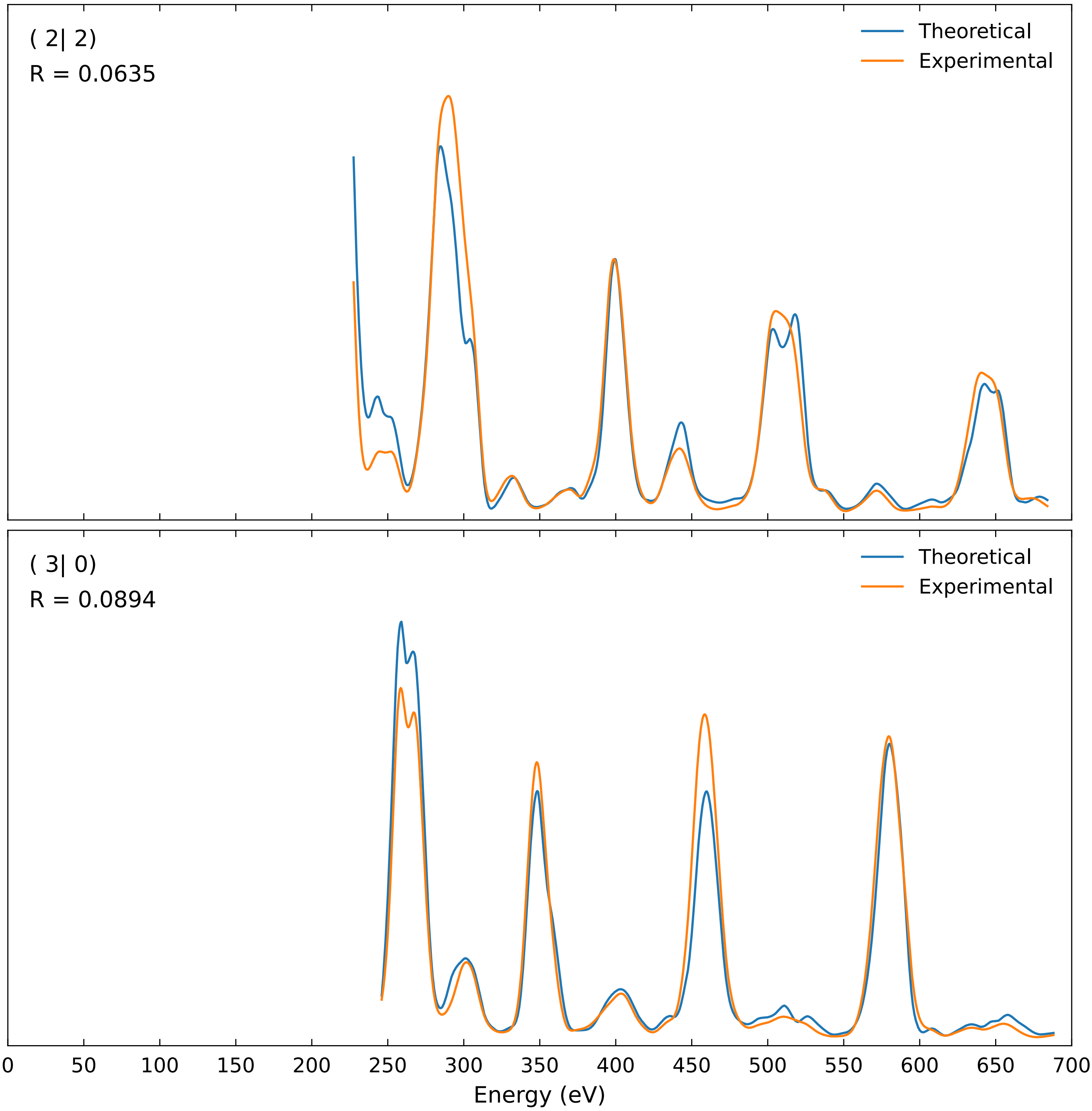


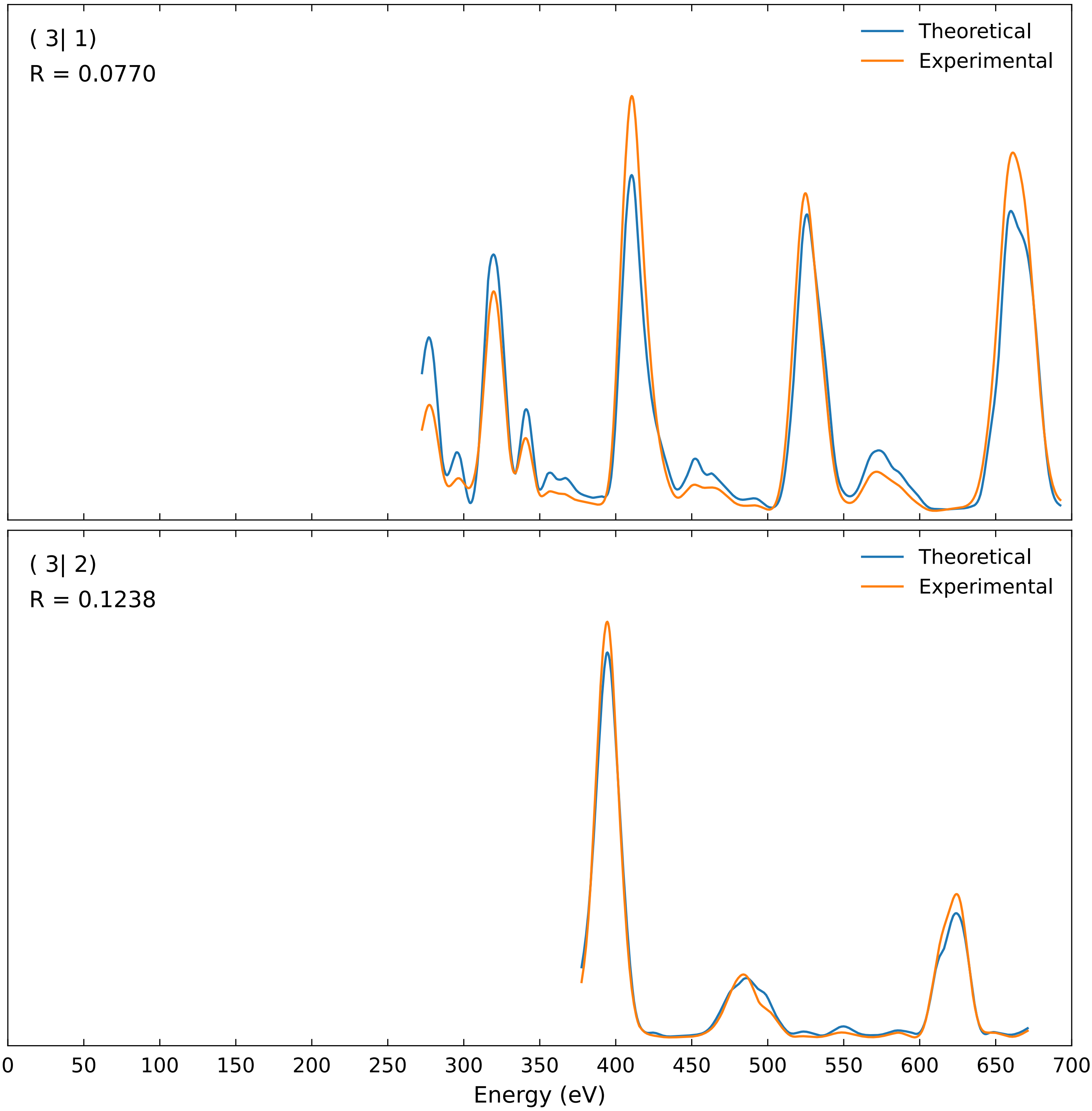
(4| 0)
R = 0.1915
Theoretical
Experimental
(4| 1)
R = 0.0375
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

Figure S4: Matching results of LEED-I(V) curves for the Ag(100)-(1×1) surface at stage D in Fig. 2a. Experimental and calculated LEED-I(V) curves are shown for all diffraction beams.




(2| 0)
R = 0.1064
Theoretical
Experimental
(2| 1)
R = 0.0790
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)


(2| 2)
R = 0.0635
Theoretical
Experimental
(3| 0)
R = 0.0894
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)


(3| 1)
R = 0.0770
Theoretical
Experimental
(3| 2)
R = 0.1238
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

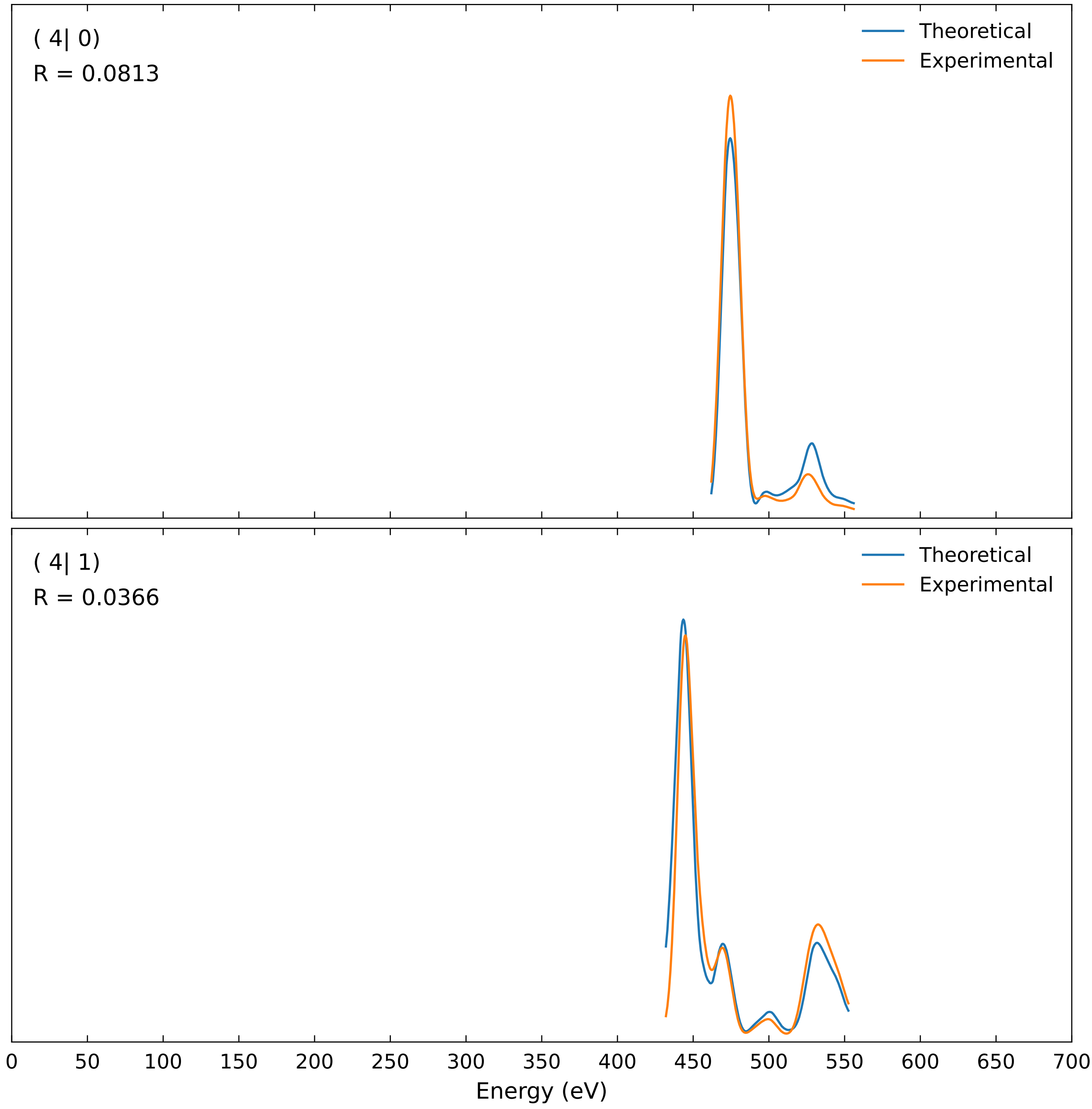
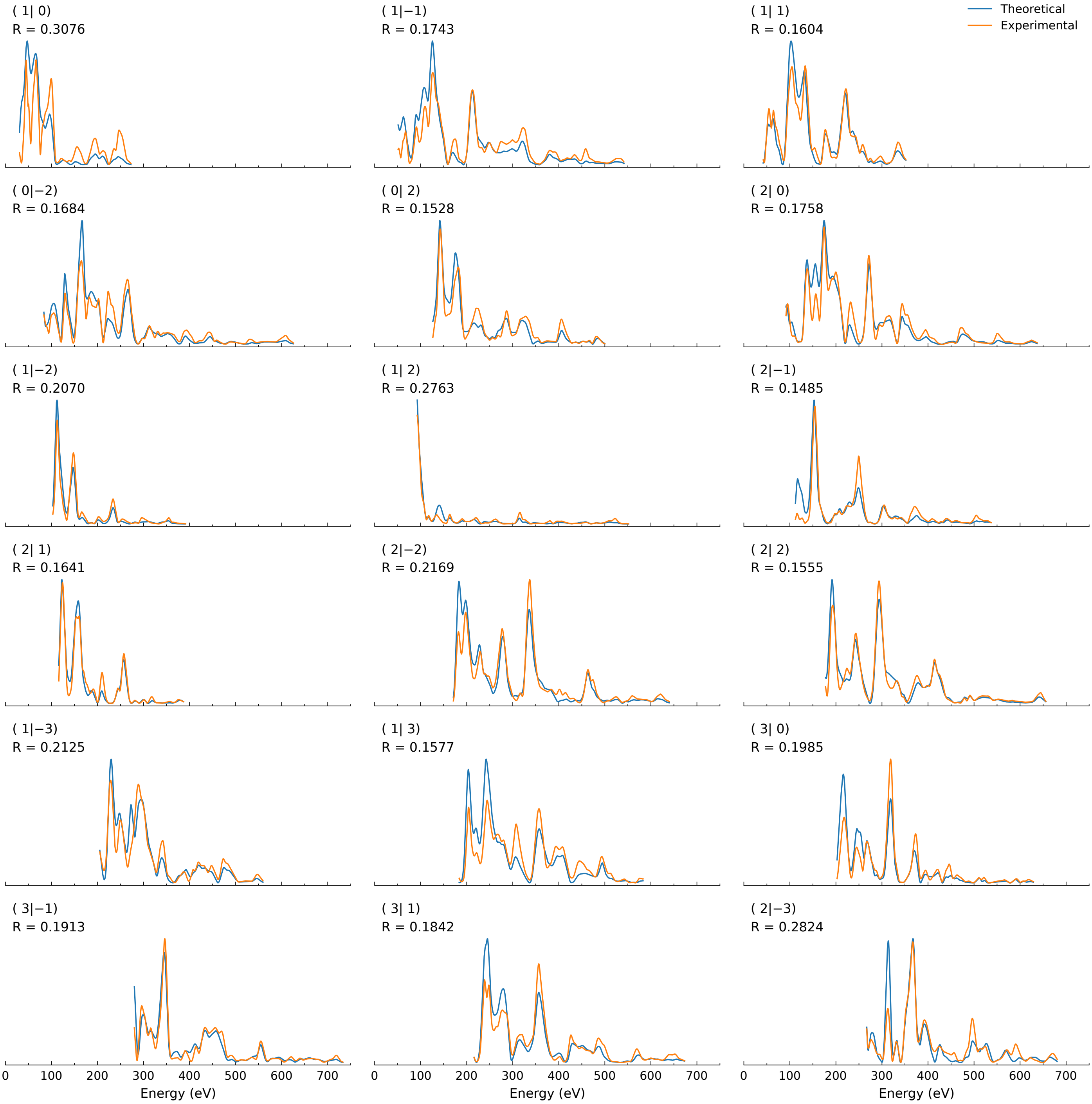
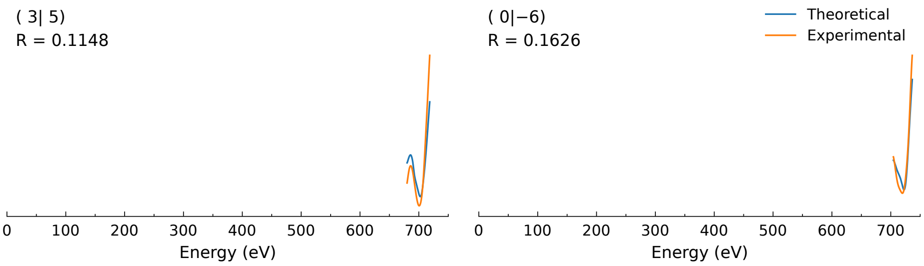

(4| 0)
R = 0.0813
Theoretical
Experimental
(4| 1)
R = 0.0366
Theoretical
Experimental
0
50
100
150
200
250
300
350
400
450
500
550
600
650
700
Energy (eV)

Figure S5: The final matching results of LEED-I(V) curves for the $Fe_2O_3(1\bar{1}02)$-(1×1) surface in Fig. 4. Experimental and calculated LEED-I(V) curves are shown for all diffraction beams.



(2| 3)
R = 0.1870
(3|−2)
R = 0.2030
(3| 2)
R = 0.1098
Theoretical
Experimental
(0|−4)
R = 0.2416
(0| 4)
R = 0.2194
(1|−4)
R = 0.2826
(1| 4)
R = 0.3581
(4| 0)
R = 0.1628
(3|−3)
R = 0.2218
(3| 3)
R = 0.2233
(4|−1)
R = 0.1543
(4| 1)
R = 0.1764
(2| 4)
R = 0.2026
(4| 2)
R = 0.0898
(1| 5)
R = 0.1854
(4|−3)
R = 0.2332
(4| 3)
R = 0.2946
(4| 4)
R = 0.1402
0
100
200
300
400
500
600
700
Energy (eV)


(3| 5)
R = 0.1148
0
100
200
300
400
500
600
700
Energy (eV)
(0|−6)
R = 0.1626
Theoretical
Experimental
0
100
200
300
400
500
600
700
Energy (eV)

References

[1] S. L. Dudarev, G. A. Botton, S. Y. Savrasov, C. J. Humphreys, and A. P. Sutton, "Electron-energy-loss spectra and the structural stability of nickel oxide: An LSDA+U study," *Physical Review B*, vol. 57, no. 3, pp. 1505–1509, Jan. 1998. [Online]. Available: https://link.aps.org/doi/10.1103/PhysRevB.57.1505

Table S3: Optimized results of $Fe_2O_3(1\bar{1}02)$-(1×1) surface

Parameters	**Value**	
	BO	**ViperLEED.calc**
R_P	0.1977	0.2109
$\mathrm{VIBROCC_{Fe}}$	0.1275 Å	0.1250 Å
$\mathrm{VIBROCC_{O}}$	0.1762 Å	0.1760 Å
θ	0.8740°	0.8538°
Atomic Coordinates		
1 Fe surf	(0.215355, 0.798869, 0.460520)	(0.218226, 0.796729, 0.460498)
2 Fe surf	(0.776472, 0.296385, 0.460520)	(0.781774, 0.296729, 0.460498)
3 Fe def	(0.736993, 0.716965, 0.413328)	(0.741063, 0.709060, 0.413316)
4 Fe def	(0.254858, 0.213301, 0.413328)	(0.258936, 0.209060, 0.413316)
5 Fe def	(0.751326, 0.291542, 0.338684)	(0.749007, 0.288387, 0.338722)
6 Fe def	(0.245944, 0.788544, 0.338684)	(0.250993, 0.788387, 0.338722)
7 Fe def	(0.245691, 0.209576, 0.286705)	(0.250000, 0.206368, 0.286789)
8 Fe def	(0.744757, 0.708996, 0.286705)	(0.750000, 0.706368, 0.286789)
9 Fe def	(0.250000, 0.790230, 0.213723)	(0.250000, 0.790230, 0.213723)
10 Fe def	(0.750000, 0.290230, 0.213723)	(0.750000, 0.290230, 0.213723)
11 Fe def	(0.750000, 0.709770, 0.161280)	(0.750000, 0.709770, 0.161280)
12 Fe def	(0.250000, 0.209770, 0.161280)	(0.250000, 0.209770, 0.161280)
13 Fe def	(0.750000, 0.290230, 0.088723)	(0.750000, 0.290230, 0.088723)
14 Fe def	(0.250000, 0.790230, 0.088723)	(0.250000, 0.790230, 0.088723)
15 Fe def	(0.250000, 0.209770, 0.036280)	(0.250000, 0.209770, 0.036280)
16 Fe def	(0.750000, 0.709770, 0.036280)	(0.750000, 0.709770, 0.036280)
17 O surf	(0.898411, 0.632551, 0.475420)	(0.905679, 0.632241, 0.475338)
18 O surf	(0.087321, 0.145573, 0.475420)	(0.094321, 0.132241, 0.475338)
19 O def	(0.449518, 0.500102, 0.441663)	(0.443607, 0.500611, 0.441913)
20 O def	(0.549706, 0.014055, 0.441663)	(0.556393, 0.000611, 0.441913)
21 O def	(0.903595, 0.336538, 0.397949)	(0.896742, 0.342033, 0.398137)
22 O def	(0.088396, 0.840688, 0.397949)	(0.103258, 0.842033, 0.398137)
23 O def	(0.400037, 0.156677, 0.352109)	(0.403693, 0.154376, 0.352035)
24 O def	(0.592263, 0.656793, 0.352109)	(0.596307, 0.654376, 0.352035)
25 O def	(0.941174, 0.998375, 0.313209)	(0.943607, 0.000378, 0.313179)
26 O def	(0.061625, 0.499275, 0.313209)	(0.056393, 0.500378, 0.313179)
27 O def	(0.387653, 0.846908, 0.274741)	(0.398728, 0.845552, 0.274496)
28 O def	(0.595992, 0.343286, 0.274741)	(0.601272, 0.345552, 0.274496)
29 O def	(0.902700, 0.652700, 0.225677)	(0.902700, 0.652700, 0.225677)
30 O def	(0.097300, 0.152700, 0.225677)	(0.097300, 0.152700, 0.225677)
31 O def	(0.444600, 0.500000, 0.187500)	(0.444600, 0.500000, 0.187500)
32 O def	(0.555400, 0.000000, 0.187500)	(0.555400, 0.000000, 0.187500)
33 O def	(0.902700, 0.347300, 0.149325)	(0.902700, 0.347300, 0.149325)
34 O def	(0.097300, 0.847300, 0.149325)	(0.097300, 0.847300, 0.149325)
35 O def	(0.402700, 0.152700, 0.100677)	(0.402700, 0.152700, 0.100677)
36 O def	(0.597300, 0.652700, 0.100677)	(0.597300, 0.652700, 0.100677)
37 O def	(0.944600, 0.000000, 0.062500)	(0.944600, 0.000000, 0.062500)
38 O def	(0.055400, 0.500000, 0.062500)	(0.055400, 0.500000, 0.062500)
39 O def	(0.402700, 0.847300, 0.024326)	(0.402700, 0.847300, 0.024326)
40 O def	(0.597300, 0.347300, 0.024326)	(0.597300, 0.347300, 0.024326)