Data set	Molecule A	Molecule B	Mean in nm	Min in nm	Max in nm	Median in nm	Std.Dev in nm
1	LAG	TCR	45.73	0.87	310.02	37.01	32.04
2	LAG	TCR	44.28	0.87	326.28	34.89	33.09
3	LAG	TCR	45.45	0.81	377.42	35.46	35.4
4	LAG	TCR	46.88	0.86	346.97	38.74	31.95
5	LAG	TCR	51.49	0.81	376.87	42.06	36.04
6	LAG	TCR	56.44	0.32	366.51	46.81	37.86
7	LAG	TCR	49.3	0.21	354.65	39.31	36.69
8	LAG	TCR	49.59	0.71	358.14	39.73	35.54
9	LAG	TCR	50.25	0.71	358.14	40.27	35.89
10	LAG	TCR	54.68	0.71	374.93	44.99	37.18
11	LAG	TCR	52.37	0.47	357.74	42.87	36.83
12	LAG	TCR	49.08	0.62	404.48	39.73	34.92
13	LAG	TCR	57.62	0.58	390.11	49.2	36.61
14	LAG	TCR	47.25	0.48	350.19	38.2	34.7
15	LAG	TCR	47.98	0.48	350.19	38.92	34.33
1	LAG	CD4	113.68	1.54	576.62	93.11	76.93
2	LAG	CD4	102.9	1.77	539.76	84.51	68.5
3	LAG	CD4	108.36	1.28	542.29	85.34	77.67
4	LAG	CD4	103.56	1.17	534.2	88.6	66.33
5	LAG	CD4	126.46	1.42	728.05	103.31	94.07
6	LAG	CD4	115.32	1.7	511.31	100.65	69.36
7	LAG	CD4	118.45	2.37	558.55	96.48	80.66
8	LAG	CD4	122.43	1.1	738.23	98.87	88.75
9	LAG	CD4	117.56	1.1	783.24	96.99	82.13
10	LAG	CD4	133.82	1.01	638.41	112.97	88.39
11	LAG	CD4	129.06	1.09	594.81	110.58	84.13
12	LAG	CD4	131.12	1.51	771.74	107.88	90.56
13	LAG	CD4	129.53	1.25	924.91	116.32	74.27
14	LAG	CD4	146.42	0.93	592.13	128.15	89.87
15	LAG	CD4	148.53	0.93	797.57	129.33	92.9

Supplementary Table 1: Summary of the shortest distance from LAG3 to TCR and CD4.

Data Set	Percentage of excess zero counts of LAG3 in the vicinity of TCR	Mean count of the number of LAG3 in the vicinity of TCR	Percentage of excess zero counts of CD4 in the vicinity of TCR	Mean count of the number of CD4 in the vicinity of TCR
1	38.44	12.4273	79.74%	1.028348
2	38.82	16.01264	79.08%	1.246302
3	53.95	17.06672	83.93%	2.179631
4	46.27	11.62417	82.00%	0.883695
5	40.76	13.07961	85.00%	1.31678
6	43.39	8.998224	84.27%	0.911355
7	52.59	16.4538	82.51%	0.941975
8	62.72	14.37803	86.67%	0.891791
9	60.8	14.48868	84.78%	0.855928
10	63.36	10.76584	88.12%	0.832506
11	51.08	11.69271	83.40%	1.845687
12	63.33	13.39716	88.45%	1.162308
13	45.5	6.872539	90.93%	7.604515
14	60.34	16.33437	86.97%	0.927899
15	58.45	15.95207	86.80%	0.925333

Supplementary Table 2: Percentage of excess zero counts and mean count of molecules of LAG3 in the vicinity of TCR and CD4 using zero-inflated Poisson regression

LAG3 peptide	N	ΔH (kcal/mol)	-TΔS (kcal/mol/deg)	ΔG (kcal/mol)	K_d (μM)
Mouse EP	1.85 ± 0.677	0.169 ± .008	-6.41 ± 0.212	-6.24 ± 0.205	19.5 ± 6.64
Human EP	2.21 ± 0.133	0.805 ± 0.067	-6.49 ± 0.075	-5.68 ± 0.117	49.5 ± 8.95
Mouse DP	1.95 ± 0.274	0.584 ± 0.005	-6.11 ± 0.089	-5.53 ± 0.891	64.4 ± 10.7
Mouse QP	n.d.	n.d.	n.d.	n.d.	n.d.
Mouse EG	1.51 ± 0.035	0.256 ± 30.10	-6.42 ± 0.144	-6.17 ± 0.174	22.1 ± 6.34

Supplementary Table 3: The disordered cytoplasmic tail of LAG3 binds Zn^{2+} weakly, in an entropically driven thermodynamic process. ITC curves at 15°C of Zn^{2+} titrated into 75 μM LAG3 peptides, in 10 mM Tris pH 7.0 buffer show that acidic residues are required for metal binding.