Assessing Nickel Titanium Binary Systems Using Structural Search Methods and Ab Initio Calculations

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Crystallographic, Elastic, and Thermal data

Table S 1: Crystallographic information for structures found on the convex hull with MHM. In addition, low-energy structures from OQMD,[2] Materials Project,[1] and experiment are listed in the table. Experimental values are in bold.

Composition	Atoms per unit cell	Space Group	a (Å)	b (Å)	c (Å)	α	β	γ
$NiTi_2$	6	I4/mmm	5.30	5.30	5.30	147.0	147.0	47.3
$NiTi_2$ [1]	24	Fd-3m	7.97	7.97	7.97	60.0	60.0	60.0
$NiTi_2$ [1*]	24	Fd-3m	7.97	7.97	7.97	60.0	60.0	60.0
$NiTi_2$ [2]	**24**	**Fd-3m**	**7.95**	**7.95**	**7.95**	**60.0**	**60.0**	**60.0**
$NiTi_2$ [3]	**24**	**Fd-3m**	**7.95**	**7.95**	**7.95**	**60.0**	**60.0**	**60.0**
$NiTi_2$ [4]	**24**	**Fd-3m**	**7.97**	**7.97**	**7.97**	**60.0**	**60.0**	**60.0**
NiTi	4	$P2_1/m$	2.92	4.01	4.89	90.0	106.3	90.0
NiTi	4	$P2_1/m$	2.93	4.06	4.75	90.0	100.9	90.0
NiTi [2]	4	$P2_1/m$	2.91	4.03	4.70	90.0	98.4	90.0
NiTi [2]	**4**	**$P2_1/m$**	**2.84**	**4.13**	**4.61**	**90.0**	**95.5**	**90.0**
NiTi [2]	**4**	**$P2_1/m$**	**2.75**	**4.22**	**4.63**	**90.0**	**92.1**	**90.0**
NiTi [5]	**4**	**$P2_1/m$**	**2.90**	**4.11**	**4.65**	**90.0**	**97.8**	**90.0**
NiTi	4	Cmcm	4.91	4.91	4.01	90.0	90.0	145.2
NiTi [1]	**4**	**Cmcm**	**4.92**	**4.92**	**4.02**	**90.0**	**90.0**	**145.5**
NiTi	8	Pnma	5.20	5.20	4.20	90.0	90.0	90.0
Ni_3Ti	8	$P6_3/mmc$	5.11	5.11	4.17	90.0	90.0	120.0
Ni_3Ti [1]	16	$P6_3/mmc$	5.10	5.10	8.31	90.0	90.0	120
Ni_3Ti [2]	8	$P6_3/mmc$	5.10	5.10	4.15	90.0	90.0	120.0
Ni_3Ti [2]	**16**	**$P6_3/mmc$**	**5.08**	**5.08**	**8.29**	**90.0**	**90.0**	**120.0**
Ni_3Ti	4	Pm-3m	3.61	3.61	3.61	90.0	90.0	90.0
Ni_3Ti [1]	4	Pm-3m	3.94	3.94	3.94	90.0	90.0	90.0
Ni_3Ti [2]	4	Pm-3m	3.60	3.60	3.60	90.0	90.0	90.0
Ni_5Ti	6	Cmmm	3.57	3.60	5.63	90.0	90.0	90.0

Table S 2: Elastic properties for convex hull structures found with MHM. B, E, and S stand for the bulk, Young's, and shear moduli, respectively. ν is the Poisson Ratio. v_l, v_t, and v_{avg} are the longiudal, transverse, and the average velocity. The elastic constant values are the averages of the Voigt and Reuss methods for determining the values of each elastic property from the elastic constants. Other strong materials are included for comparison. Two metastable structures with unique properties are included at the end of the table for comparison.

Composition	Space Group	B (GPa)	E (GPa)	S (GPa)	ν (GPa)	B/S	v_l (m/s)	v_t (m/s)	v_{avg} (m/s)	P-wave modulus	Universal anisotropy	Log-Euclidean anisotropy
Ti	P6/mmm	113.5	157.0	61.9	0.2	1.8	6495	3649	4060	196.01	0.232	0.234
$NiTi_2$	I4/mmm	145.2	170.2	65.2	0.3	2.2	6307	3343	3736	232.22	0.421	0.416
$NiTi_2$	Fd-3m	143.0	98.0	35.9	0.361	3.97	4866	2482	2781	191.00	2.808	2.296
$NiTi_2$[6]	Fd-3m	146.1	44.4	49.4	0.348	2.96						
$NiTi_2$[1]	Fd-3m	141.7	99.1	35.8	0.383	3.96						
NiTi	Pnma	117.1	128.1	48.6	0.3	2.4	5296	2738	3066	181.92	2.476	2.072
NiTi	$P2_1/m$	158.0	111.1	40.8	0.4	3.9	5751	2521	2846	212.45	8.350	5.057
NiTi	Pm-3m	164.54	92.28	32.85	0.407	5.00	5653	2244	2543	208.3	1.716	1.520
Ni_3Ti	$P6_3/mmc$	196.1	233.2	89.6	0.3	2.2	6323	3369	3764	315.53	0.299	0.299
Ni_5Ti	Cmmm	199.2	230.6	88.3	0.3	2.3	6199	3271	3657	316.90	0.886	0.840
Ni	Fm-3m	207.2	211.9	80.0	0.3	2.6	5901	2976	3338	313.67	1.418	1.286
W[7]	Im-3m	304.0	380.5	147.5	0.3	2.06						
Stainless Steel[8]	fcc	152	198	77	0.28	1.98						
Stainless Steel[9]	fcc	142-186	195-208	75-82	0.27-0.32	1.88-2.45						
$NiTi_2$	C2/m	140.9	75.7	27.0	0.4	5.22	5556	2170	2460	176.91	3.993	3.023
NiTi	P2/m	155.1	97.9	35.1	0.4	4.41	5447	2166	2454	193.49	3.876	2.956

Table S 3: Thermodynamic properties of Ni_xTi_{1-x}. C_v is the specific heat with constant volume. The values above above the dividing line are from first principles studies at 300K. The values below the line are experimental reported values for debye temperature.

Composition	Space Group	C_v(J/mol K)	Debye Temp.(K)
$NiTi_2$	I4/mmm	138.78	441.82
$NiTi_2$	Fd-3m	562.73 (519.34[6])	217.92 (411.5[6])
NiTi	Pnma	185.69	392.61
NiTi	$P2_1/m$	92.73	620.79
NiTi	Cmcm	92.67	762.65
Ni_5Ti	Cmmm	139.66	899.05
Ni_3Ti	$P6_3/mmc$	183.30	510.17

Convex Hull Phonons

The phonon spectra for the convex hull structures are displayed in Figures S1,S2,S3 and S4.

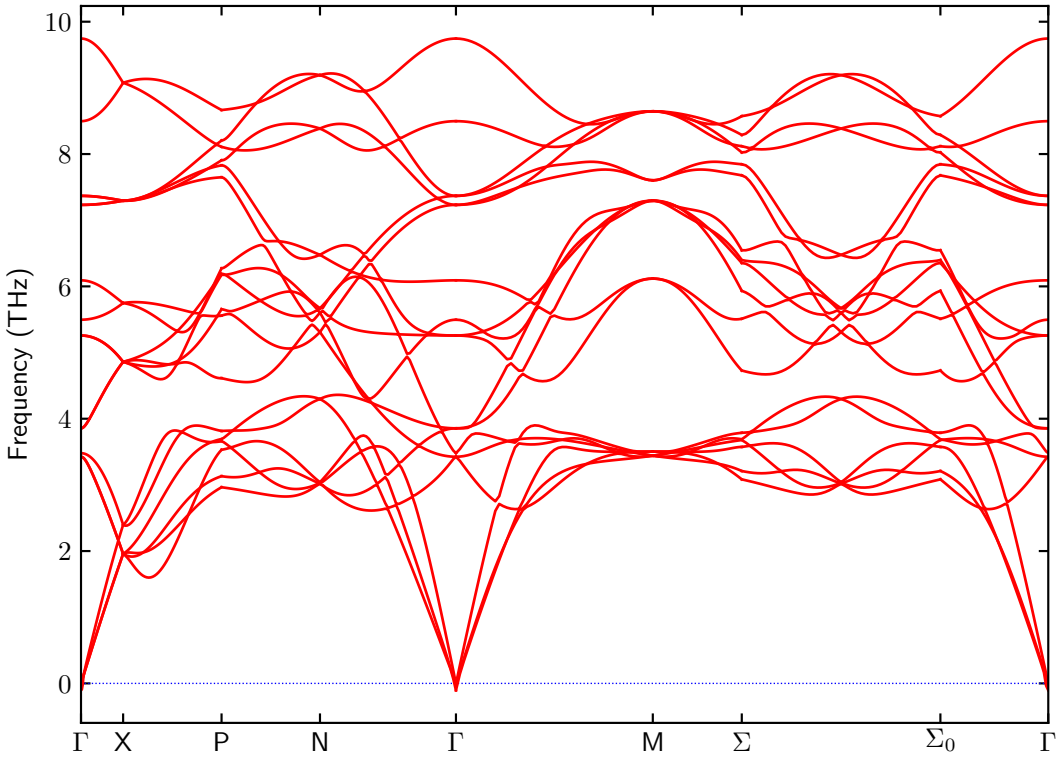


Figure S 1: Phonons for the structure on the convex hull for $NiTi_2$ with space group *I4/mmm*

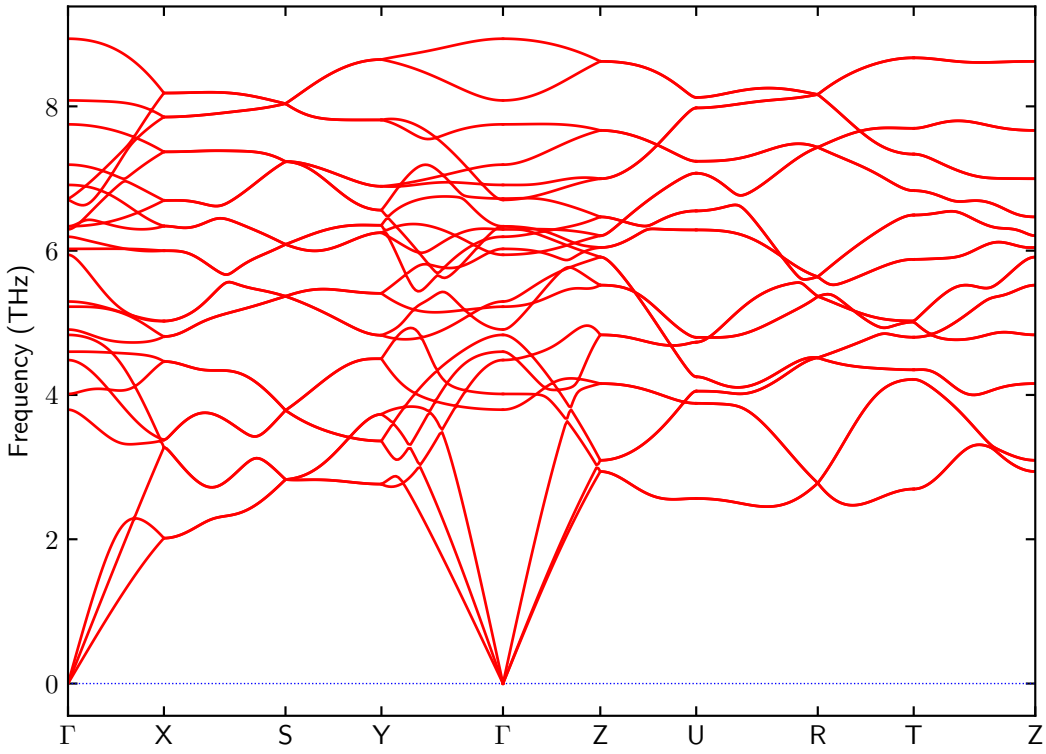


Figure S 2: Phonons for the structure on the convex hull for NiTi with space group *Pnma*

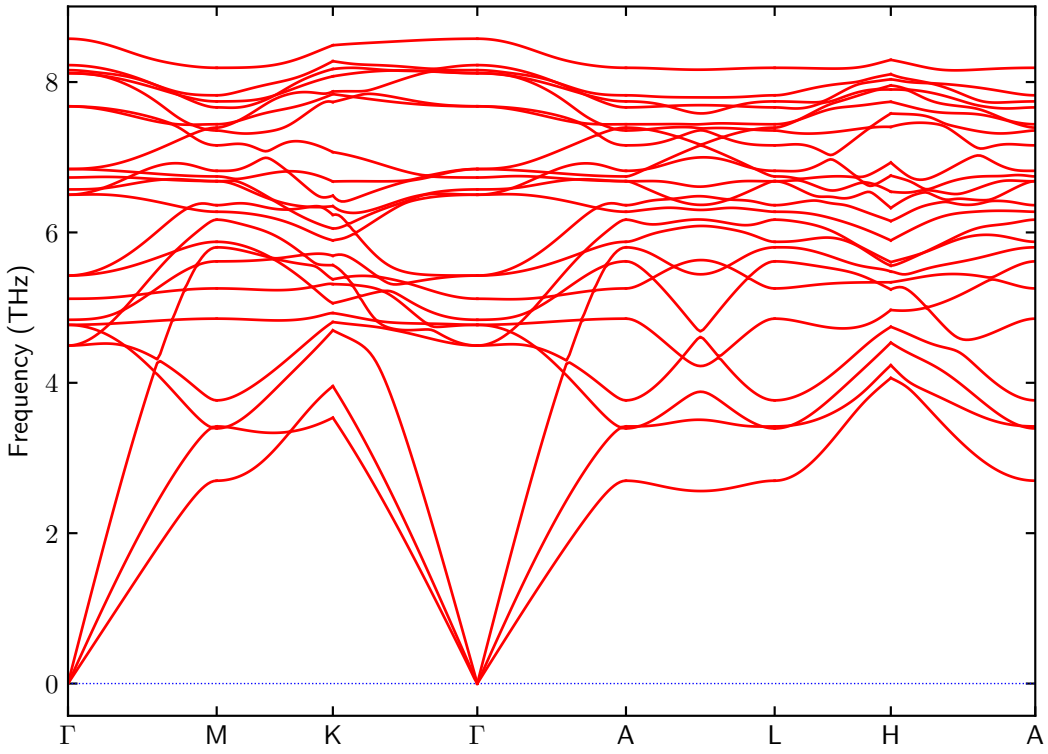


Figure S 3: Phonons for the structure on the convex hull for Ni_3Ti with space group *P6$_3$/mmc*

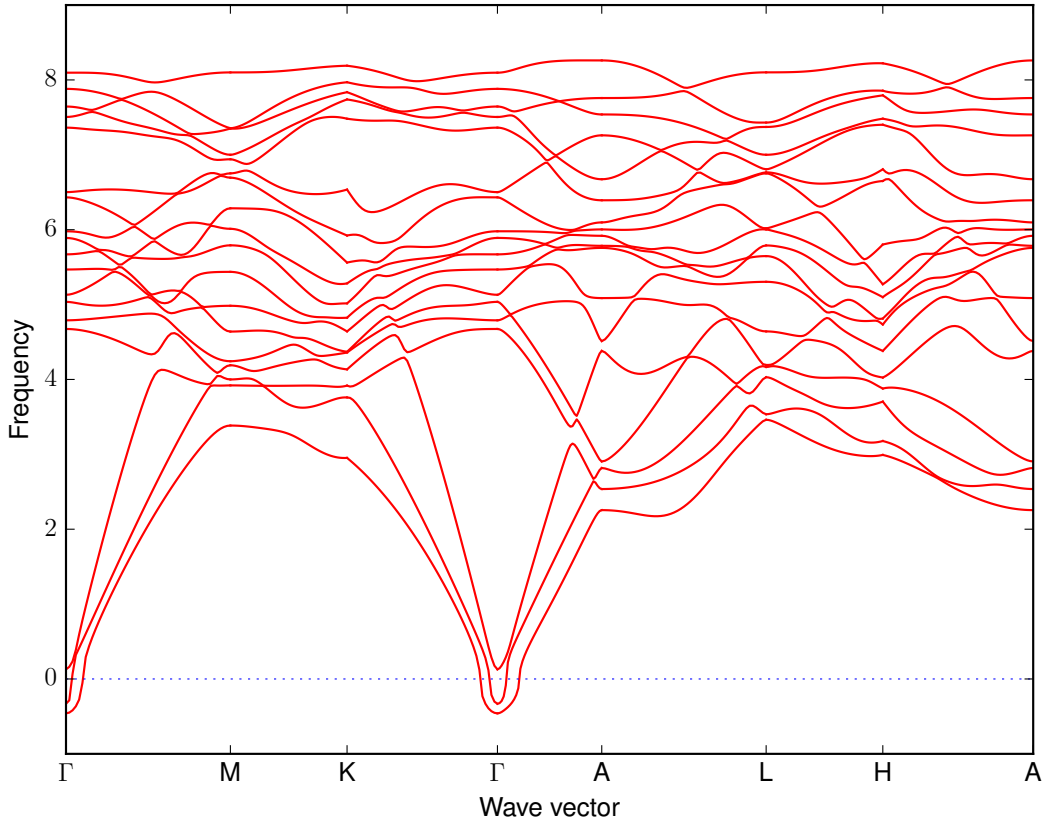


Figure S 4: Phonons for the structure on the convex hull for Ni_5Ti with space group *Cmmm*

Phonons for Metastable Structures

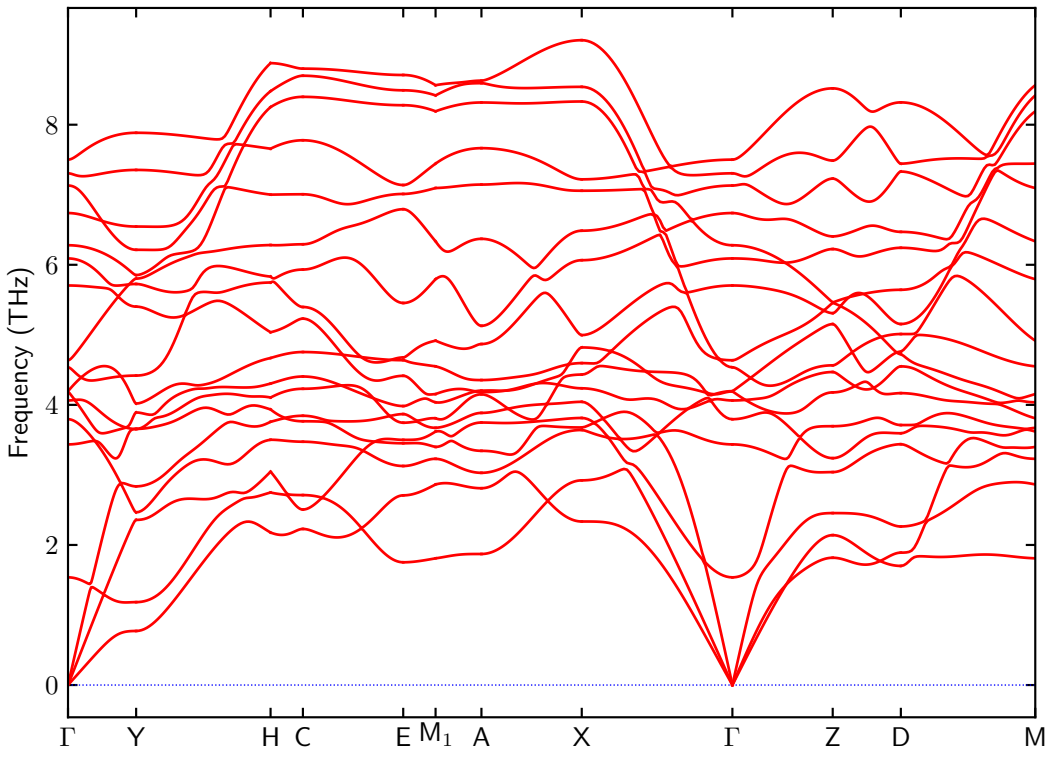


Figure S 5: Phonons for NiTi with space group *P2/m*

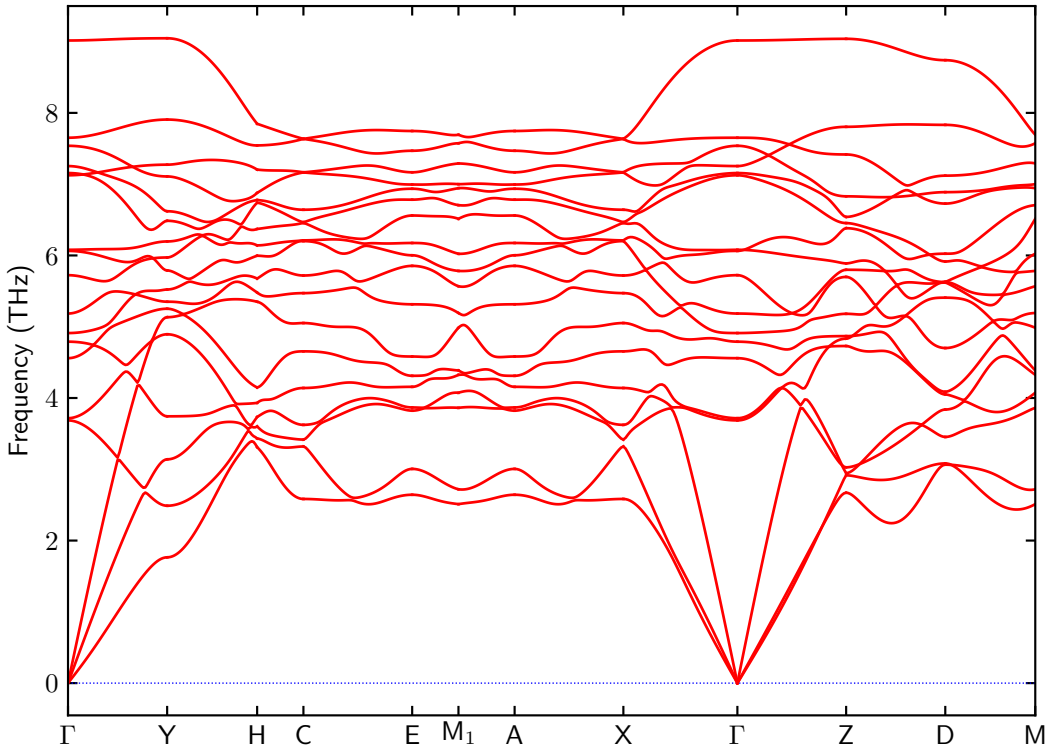
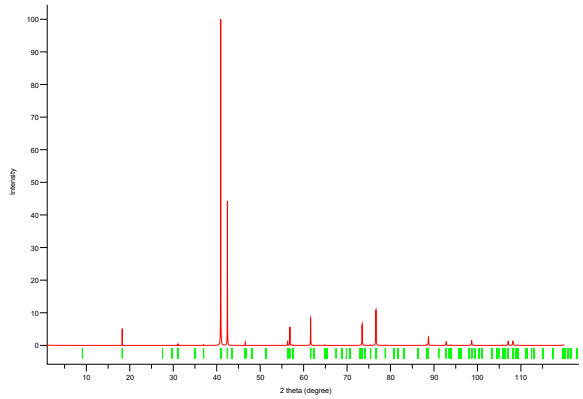
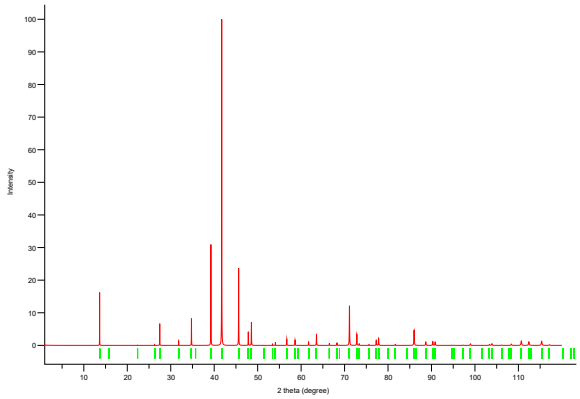


Figure S 6: Phonons for $NiTi_2$ with space group *C2/m*

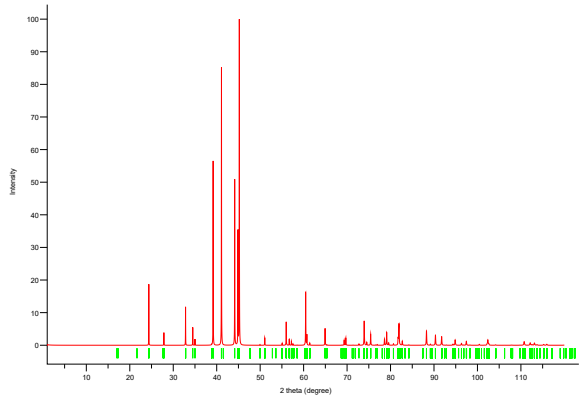
XRD



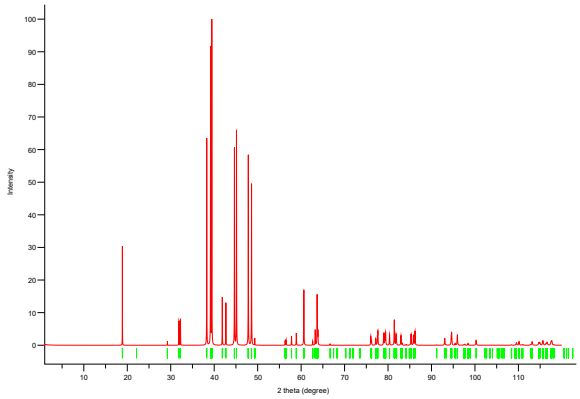
(a) $NiTi_2$ Space Group *I4/mmm*



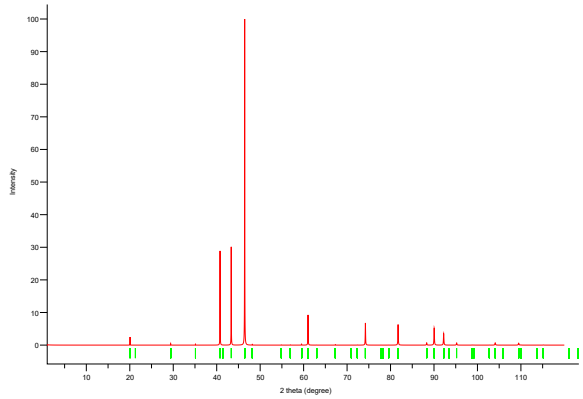
(b) $NiTi_2$ Space Group *Fd-3m*



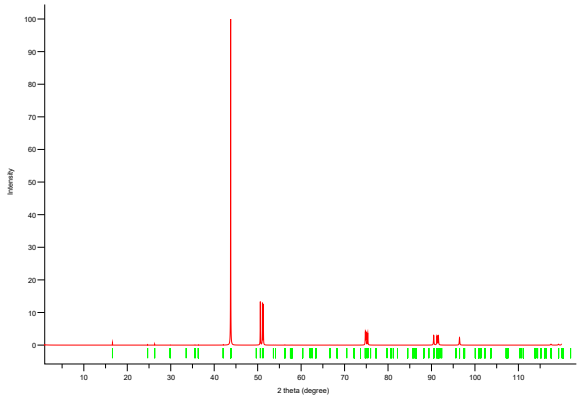
(c) NiTi Space Group *Pnma*



(d) NiTi Space Group *P2/m*



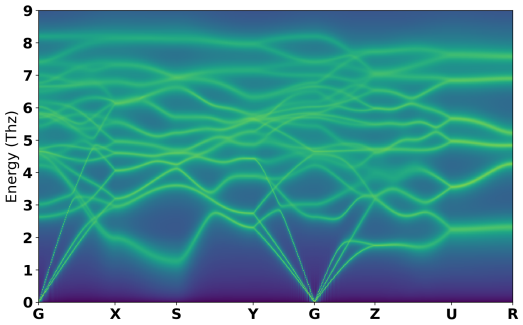
(e) Ni_3Ti Space Group *P6$_3$/mmc*



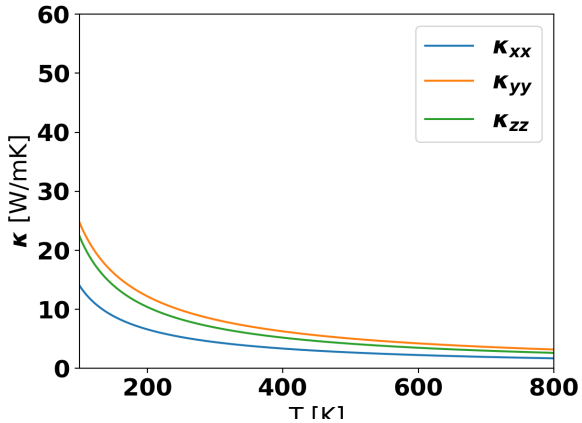
(f) Ni_5Ti Space Group *Cmmm*

Figure S 7: X-ray diffraction plots for structures reported in the paper. These plots were generated using VESTA and a Cu K-alpha X-ray ($\lambda = 1.5406\mathring{A}$) with relative intensity 1.

SQE plots and thermal conductivity



(a) NiTi-62 Spectral phonon bands



(b) NiTi-62 Thermal Conductivity

Figure S 8: Effect of anharmonicity for NiTi. Spectral Phonon Dispersion plot and Thermal conductivity temperature dependence

CIF files

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Ti Ti3 1 0.000000 0.000000 0.157567 1

Ti Ti4 1 0.500000 0.500000 0.342433 1

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Ni Ni5 1 0.839482 0.481553 0.839482 1

Ni Ni6 1 0.481553 0.839482 0.839482 1

Ni Ni7 1 0.839482 0.839482 0.839482 1

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Ti Ti9 1 0.063253 0.436747 0.063253 1

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Ni Ni4 1 0.324341 0.000000 0.162173 1

Ni Ni5 1 0.162172 0.500000 0.837832 1

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_atom_site_type_symbol

_atom_site_label

_atom_site_symmetry_multiplicity

_atom_site_fract_x

_atom_site_fract_y

_atom_site_fract_z

_atom_site_occupancy

Ni Ni0 1 0.169850 0.500000 0.339701 1

Ni Ni1 1 0.830150 0.500000 0.660299 1

Ni Ni2 1 0.500000 0.500000 0.000000 1

Ni Ni3 1 0.666738 0.000000 0.333475 1

Ni Ni4 1 0.333262 0.000000 0.666525 1

Ti Ti5 1 0.000000 0.000000 0.000000 1

$NiTi_2$ - space group 12 - C2/m

data_Ti2Ni

_symmetry_space_group_name_H-M ”P1”

_cell_length_a 4.619834

_cell_length_b 4.893469

_cell_length_c 5.117731

_cell_angle_alpha 61.439286

_cell_angle_beta 102.756527

_cell_angle_gamma 117.506927

loop_

_symmetry_equiv_pos_site_id

_symmetry_equiv_pos_as_xyz

1 x,y,z

loop_

_atom_site_label

_atom_site_type_symbol

_atom_site_fract_x

_atom_site_fract_y

_atom_site_fract_z

_atom_site_occupancy

Ni1 Ni 0.50000 0.25119 0.50762 1.0000

Ni2 Ni 0.50000 0.74881 0.49238 1.0000

Ti1 Ti 0.30987 0.17306 0.03000 1.0000

Ti2 Ti 0.69013 0.82694 0.03000 1.0000

Ti3 Ti 0.00000 0.33778 0.30444 1.0000

Ti4 Ti 0.00000 0.66222 0.69556 1.0000

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