Supplementary Material for

Synthesis, Structure, Vibrational Spectra and Thermal Analysis of Isoniazid-Succinic Acid Cocrystal

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Table S1. Crystal data, data collection and structure refinement of INH-SUC cocrystal.

Moiety formula	0.5($C_4H_6O_4$), ($C_6H_7N_3O$)
Chemical formula	$C_8H_{10}N_3O_3$
Formula weight	196,19
Crystal system	Monoclinic
Space group	$P2_1/n$ (nº 14)
Crystal form, colour	Plate, colourless
Crystal size/ mm^3	0.1 × 0.5 × 0.5
a/ Å	7.0035(4)
b/ Å	19.5966(12)
c/ Å	7.2939(4)
α/ °	90
β/ °	114.589(2)
γ/ °	90
Z, Z'	4, 1
V/ $Å^3$	910.27(9)
Density (calc., g cm^{-3})	1.432
Absorption coefficient μ/ mm^{-1}	0.112
F(000)	412.0
T/ K	303.0(5)
Radiation type	MoK_α (λ = 0.71073 Å)
2θ range for data collection/ º	6.486, 49.990
Index ranges	$-8 \leq h \leq 4$, $-23 \leq k \leq 23$, $-4 \leq l \leq 8$
Reflections collected	3115
Unique reflections ($F_o > 4\sigma(F_o)$)	1601
$R_{(int)}$, $R_{(\sigma)}$,	0.0800, 0.0883
T_{min}, T_{max}	0.946, 0.989
Data completeness	0.996
Refinement method	Full-matrix least-squares on F^2
Nref, Npar	1601, 141
R_1 (reflections)	0.0644 (1226)
wR_2(reflections)	0.1230 (1601)
GoF	1.104
Min., Max. Resd. Dens./ *e* A^{-3}	–0.196, 0.234

Table S2 - Final coordinates (Å) and equivalent isotropic displacement parameters (Å^2) of the non-hydrogen atoms.

Atom	*x*	*y*	*z*	U_{eq}
O8	0.9938(2)	0.60225(8)	0.5937(3)	0.0510(7)
N4	0.5411(3)	0.80554(9)	0.5358(3)	0.0418(7)
N9	0.6815(3)	0.55323(9)	0.5337(3)	0.0412(7)
N10	0.7576(3)	0.48530(10)	0.5687(4)	0.0436(8)
C1	0.7032(3)	0.67570(11)	0.5419(3)	0.0331(7)
C2	0.8044(4)	0.73286(11)	0.5162(4)	0.0419(8)
C3	0.7185(4)	0.79674(12)	0.5134(4)	0.0446(8)
O3A	0.1140(3)	0.87802(9)	0.5362(4)	0.0698(8)
O4A	0.3913(2)	0.92938(8)	0.5289(3)	0.0500(6)
C5	0.4422(4)	0.75032(12)	0.5594(4)	0.0426(8)
C6	0.5173(3)	0.68494(11)	0.5630(4)	0.0404(8)
C7	0.8063(3)	0.60715(11)	0.5577(3)	0.0357(7)
C1A	0.1093(3)	0.99963(11)	0.5034(4)	0.0386(8)
C2A	0.2016(3)	0.92934(11)	0.5245(3)	0.0362(7)

U_{eq} = 1/3 of the trace of the orthogonalized U tensor.

Table S3 - Hydrogen atom positions (Å) and isotropic displacement parameters (Å^2).

Atom	*x*	*y*	*z*	U_{iso}
H11	0.92922	0.72855	0.50087	0.0500
H12	0.78759	0.83492	0.49525	0.0540
H13	0.31750	0.75609	0.57408	0.0510
H14	0.44393	0.64759	0.57933	0.0480
H15	0.541(4)	0.5574(12)	0.498(4)	0.049(7)
H16	0.827(5)	0.4762(15)	0.489(4)	0.066(9)
H17	0.855(4)	0.4832(12)	0.697(4)	0.046(7)
H5A	0.43335	0.88999	0.53608	0.0750
H6A	0.20045	1.02734	0.61604	0.0460
H7A	0.10502	1.02025	0.38087	0.0460

The temperature factor has the form of Exp(-*T*), where $T = 8\pi^2 U (\mathrm{Sin}(\theta)/\lambda)^2$ for isotropic atoms.

Table S4 - Anisotropic displacement parameters (Å^2).

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O8	0.0353(10)	0.0417(10)	0.0824(14)	–0.0023(9)	0.0308(9)	0.0042(7)
N4	0.0366(11)	0.0337(10)	0.0562(13)	0.0005(9)	0.0205(10)	0.0019(8)
N9	0.0303(11)	0.0314(10)	0.0584(13)	0.0016(9)	0.0149(10)	0.0046(8)
N10	0.0373(12)	0.0325(11)	0.0617(16)	0.0011(10)	0.0214(12)	0.0075(9)
C1	0.0299(12)	0.0329(11)	0.0362(12)	–0.0012(9)	0.0134(10)	0.0018(9)
C2	0.0319(12)	0.0410(13)	0.0553(15)	0.0015(11)	0.0207(12)	–0.0004(10)
C3	0.0368(13)	0.0341(12)	0.0648(17)	0.0044(11)	0.0229(12)	–0.0036(10)
O3A	0.0563(12)	0.0350(10)	0.135(2)	0.0032(11)	0.0566(13)	–0.0008(8)
O4A	0.0396(10)	0.0323(9)	0.0856(14)	–0.0002(9)	0.0334(10)	0.0021(7)
C5	0.0347(13)	0.0411(13)	0.0569(16)	0.0016(11)	0.0238(12)	0.0037(10)
C6	0.0356(13)	0.0340(12)	0.0574(15)	0.0024(11)	0.0251(12)	–0.0012(10)
C7	0.0342(12)	0.0358(12)	0.0379(13)	0.0014(10)	0.0157(10)	0.0025(10)
C1A	0.0405(13)	0.0324(12)	0.0463(14)	0.0005(10)	0.0214(11)	0.0017(10)
C2A	0.0338(12)	0.0346(12)	0.0418(13)	–0.0028(10)	0.0174(11)	–0.0001(10)

The temperature factor has the form of Exp(–*T*) where $T = 2\pi^2 \Sigma_{ij} (h_i h_j U_{ij} A_i^* A_j^*)$, A_i^* being the reciprocal axial lengths and h_i the reflection indices.

Table S5 - Bond distances (Å)

O8-C7	1.231(3)	N9-H15	0.91(3)
N4-C3	1.330(4)	N10-H17	0.90(3)
N4-C5	1.334(3)	N10-H16	0.92(3)
N9-N10	1.417(3)	C2-H11	0.9300
N9-C7	1.336(3)	C3-H12	0.9300
C1-C2	1.379(3)	O4A-H5A	0.8200
C1-C6	1.385(3)	C5-H13	0.9300
C1-C7	1.507(3)	C6-H14	0.9300
C2-C3	1.385(3)	C1A-C2A	1.502(3)
O3A-C2A	1.199(3)	C1A-C1A_a	1.511(3)
O4A-C2A	1.316(3)	C1A-H6A	0.9700
C5-C6	1.381(3)	C1A-H7A	0.9700

Table S6 - Bond Angles (º)

C3-N4-C5	118.2(2)	C3-C2-H11	120.00
N10-N9-C7	122.9(2)	N4-C3-H12	119.00
C2-C1-C6	118.0(2)	C2-C3-H12	119.00
C2-C1-C7	118.5(2)	C2A-O4A-H5A	109.00
C6-C1-C7	123.4(2)	N4-C5-H13	119.00
C1-C2-C3	119.4(3)	C6-C5-H13	119.00
N4-C3-C2	122.5(2)	C5-C6-H14	120.00
N4-C5-C6	122.8(3)	C1-C6-H14	120.00
C1-C6-C5	119.2(2)	C1A_a-C1A-C2A	113.52(19)
O8-C7-N9	123.2(2)	O3A-C2A-C1A	124.4(2)
O8-C7-C1	121.4(2)	O4A-C2A-C1A	112.90(19)
N9-C7-C1	115.4(2)	O3A-C2A-O4A	122.7(2)
N10-N9-H15	114.6(15)	C2A-C1A-H6A	109.00
C7-N9-H15	122.5(15)	C2A-C1A-H7A	109.00
N9-N10-H16	109.1(18)	H6A-C1A-H7A	108.00
N9-N10-H17	107.2(15)	C1A_a-C1A-H6A	109.00
H16-N10-H17	106(3)	C1A_a-C1A-H7A	109.00
C1-C2-H11	120.00		

Table S7 - Torsion Angles (º)

C5-N4-C3-C2	–0.8(4)
C3-N4-C5-C6	0.5(4)
N10-N9-C7-O8	7.0(3)
N10-N9-C7-C1	–171.8(2)
C6-C1-C2-C3	0.3(4)
C7-C1-C2-C3	–176.2(2)
C2-C1-C6-C5	–0.6(4)
C7-C1-C6-C5	175.7(2)
C2-C1-C7-O8	16.6(3)
C2-C1-C7-N9	–164.6(2)
C6-C1-C7-O8	–159.7(2)
C6-C1-C7-N9	19.2(3)
C1-C2-C3-N4	0.4(4)
N4-C5-C6-C1	0.2(4)
C1A_a-C1A-C2A-O3A	–0.4(4)
C1A_a-C1A-C2A-O4A	179.6(2)
C2A-C1A-C1A_a-C2A_a	–180.0(2)

Table S8 - Contact Distances (Å)

O3A	.C5	3.354(3)	N9	.N10_f	2.945(3)
O3A	.C1A_a	2.811(3)	N9	.C6	2.870(3)
O3A	.C3_k	3.139(4)	N10	.O8	2.788(3)
O4A	.C7_d	3.313(3)	N10	.O8_c	3.016(3)
O4A	.N10_l	3.139(3)	N10	.N9_f	2.945(3)
O4A	.N4	2.636(2)	N10	.O4A_b	3.139(3)
O8	.N10_c	3.016(3)	C1	.N4	2.779(3)
O8	.C2	2.830(3)	C1	.N4_h	3.409(3)
O8	.N10	2.788(3)	C1A	.O3A_a	2.811(3)
O3A	.H7A_a	2.7300	C2	.O8	2.830(3)
O3A	.H13	2.7400	C2	.C5	2.705(4)
O3A	.H5A	2.2500	C2A	.N4	3.374(3)
O3A	.H12_k	2.3400	C3	.O3A_i	3.139(4)
O3A	.H6A_a	2.7400	C3	.C6	2.709(3)
N4	.C1_d	3.409(3)	N4	.H5A	1.8200
N4	.O4A	2.636(2)	C5	.C2	2.705(4)
N4	.C2A	3.374(3)	C5	.O3A	3.354(3)
N4	.C1	2.779(3)	C6	.C3	2.709(3)
O4A	.H17_l	2.30(3)	C6	.N9	2.870(3)
O4A	.H17_d	2.89(3)	C7	.O4A_g	3.313(3)
O4A	.H6A	2.5700	N9	.H7A_e	2.9200
O4A	.H7A	2.5600	N9	.H14	2.6000
O8	.H17	2.75(3)	N9	.H15_f	2.62(3)
O8	.H16_c	2.22(3)	N10	.H15_f	2.11(3)
O8	.H11	2.5600	N10	.H14_f	2.9400
O8	.H16	2.70(3)	N10	.H7A_g	2.8900
O8	.H6A_b	2.7300	C1	.H15	2.54(2)
N9	.N9_f	3.165(3)	C1A	.H5A	3.0600
C2A	.H7A_a	2.7000	H11	.C7	2.6200
C2A	.H6A_a	2.7000	H12	.O3A_i	2.3400
C3	.H5A	2.7600	H12	.H11	2.3000
C5	.H5A	2.7400	H13	.O3A	2.7400
C6	.H15	2.56(2)	H13	.H14	2.3000
C7	.H17	2.60(2)	H14	.C7	2.7300
C7	.H11	2.6200	H14	.N9	2.6000
C7	.H14	2.7300	H14	.N10_f	2.9400
C7	.H16	2.63(3)	H14	.H13	2.3000
H5A	.O3A	2.2500	H14	.H15	2.0700
H5A	.N4	1.8200	H15	.H16	2.58(4)
H5A	.C1A	3.0600	H15	.C1	2.54(2)
H5A	.C3	2.7600	H15	.C6	2.56(2)
H5A	.C5	2.7400	H15	.H14	2.0700
H6A	.O4A	2.5700	H15	.N9_f	2.62(3)
H6A	.O8_l	2.7300	H15	.N10_f	2.11(3)
H6A	.O3A_a	2.7400	H15	.H17	2.53(4)
H6A	.C2A_a	2.7000	H15	.H15_f	2.33(3)
H6A	.H7A_a	2.3400	H16	.H15	2.58(4)
H6A	.H16_m	2.4900	H16	.O8_c	2.22(3)
H7A	.N9_n	2.9200	H16	.O8	2.70(3)
H7A	.O3A_a	2.7300	H16	.C7	2.63(3)
H7A	.C2A_a	2.7000	H16	.H16_c	2.54(5)
H7A	.H6A_a	2.3400	H16	.H6A_j	2.4900
H7A	.N10_d	2.8900	H17	.O4A_g	2.89(3)
H7A	.O4A	2.5600	H17	.O8	2.75(3)
H11	.H12	2.3000	H17	.C7	2.60(2)
H11	.O8	2.5600	H17	.H15	2.53(4)
H17	.O4A_b	2.30(3)			

Table S9 - Translation of symmetry code to equivalent positions.

a =	[3576.00] = [3_576]	= –x,2–y,1–z
b =	[2646.00] = [2_646]	= 3/2-x,–1/2+y,3/2–z
c =	[3766.00] = [3_766]	= 2–x,1–y,1–z
d =	[4464.00] = [4_575]	= –1/2+x,3/2–y,–1/2+z
e =	[2545.00] = [2_545]	= 1/2-x,–1/2+y,1/2–z
f =	[3666.00] = [3_666]	= 1–x,1–y,1–z
g =	[4565.00] = [4_676]	= 1/2+x,3/2–y,1/2+z
h =	[4565.00] = [4_676]	= 1/2+x,3/2–y,1/2+z
i =	[1655.00] = [1_655]	= 1+x,y,z
j =	[4564.00] = [4_675]	= 1/2+x,3/2–y,–1/2+z
k =	[1455.00] = [1_455]	= –1+x,y,z
l =	[2656.00] = [2_656]	= 3/2-x,1/2+y,3/2–z
m =	[4465.00] = [4_576]	= –1/2+x,3/2–y,1/2+z
n =	[2555.00] = [2_555]	= 1/2–x,1/2+y,1/2–z

Table S10 – Definition of the internal coordinates used in the normal coordinate analysis of INH.[a]

Coordinate	Definition	Approximate description
S1	ν(N10–H16) + ν(N10–H17)	ν(NH_2) s
S2	ν(N10–H16) – ν(N10–H17)	ν(NH_2) as
S3	ν(N9–N10)	ν(N–N)
S4	ν(N9–H15)	ν(N–H)
S5	ν(N9–C7)	ν(N–C)
S6	ν(C7=O8)	ν(C=O)
S7	ν(C1–C7)	ν(C–C)
S8	ν(C3–H12)	ν(C–H) 1
S9	ν(C5–H13)	ν(C–H) 2
S10	ν(C2–H11)	ν(C–H) 3
S11	ν(C6–H14)	ν(C–H) 4
S12	ν(C3–C2) + ν(N4–C3) + ν(N4–C5) + ν(C6–C5) + ν(C1–C2) + ν(C1–C6)	ν_{Ring} 1
S13	ν(C3–C2) – ν(N4–C3) + ν(N4–C5) – ν(C6–C5) + ν(C1–C2) – ν(C1–C6)	ν_{Ring} 2
S14	2ν(C3–C11) – ν(N4–C3) – ν(N4–C5) + 2ν(C6–C5) – ν(C1–C2) – ν(C1–C6)	ν_{Ring} 3
S15	ν(N4–C3) – ν(N4–C5) + ν(C1–C2) – ν(C1–C6)	ν_{Ring} 4
S16	ν(C3–C2) – 2ν(C6–C5)	ν_{Ring} 5
S17	–ν(N4–C3) – ν(N4–C5) + ν(C1–C2) + ν(C1–C6)	ν_{Ring} 6
S18	2δ(H16–H17–N10) – δ(N9–H16–N10) – δ(N9–H17–N10)	δ(NH_2)
S19	δ(N9–H16–N10) – δ(N9–H17–N10)	w(NH_2)
S20	2δ(N10–C7–N9) – δ(H15–N10–N9) – δ(H15–C7–N9)	δ(CNN)
S21	δ(H15–N10–N9) – δ(H15–C7-N9)	δ(N–H)
S22	2δ(C1–N9–C7) – δ(O8–C1–C7) – δ(O8–N9–C7)	δ(CCO)
S23	δ(O8–C1–C7) – δ(O8–N9–C7)	δ(C=O)
S24	δ(C7–C2–C1) – δ(C7–C6–C5)	w(C–$CONHNH_2$)
S25	δ(H11–C1–C2) – δ(H11–C3–C2) + δ(H12–C2–C3) – δ(H12 –N4–C3) + δ(H13–N4–C5) – δ(H13–C6–C5) + δ(H14–C5–C6) – δ(H14–C1–C6)	δ(C–H) 1
S26	δ(H11–C1–C2) – δ(H11–C3–C2) – δ(H12–C2–C3) + δ(H12 –N4–C3) + δ(H13–N4–C5) – δ(H13–C6–C5) – δ(H14–C5–C6) + δ(H14–C1–C6)	δ(C–H) 2
S27	δ(H11–C1–C2) – δ(H11–C3–C2) – δ(H12–C2–C3) + δ(H12 –N4–C3) – δ(H13–N4–C5) + δ(H13–C6–C5) + δ(H14–C5–C6) – δ(H14–C1–C6)	δ(C–H) 3
S28	δ(H11– C1– C2) – δ(H11–C3–C2) + δ(H12–C2–C3) – δ(H12 –N4–C3) – δ(H13–N4–C5) + δ(H13–C6–C5) – δ(H14–C5–C6) + δ(H14–C1–C6)	δ(C–H) 4
S29	δ(C1–C3–C2) – δ(C2–N4–C3) + δ(C5–C3–N4) – δ(C6–N4–C5) + δ(C1–C5–C6) – δ(C2–C6–C1)	δ_{Ring} 1
S30	δ(C1–C3–C2) – δ(C2–N4–C3) + δ(C6–N4–C5) – δ(C1–C5–C6)	δ_{Ring} 2
S31	δ(C1–C3–C2) + δ(C2–N4–C3) – 2δ(C5–C3–N4) + δ(C6–N4–C5) + δ(C1–C5–C6) –2δ(C2–C6–C1)	δ_{Ring} 3
S32	τ(C7–N9–N10–H17) + τ(C7–N9–N10–H16)	τ(N–N)
S33	τ(C1–C7–N9–N10)	τ(C–N)
S34	τ(C2–C1–C7–N9)	τ(C–C)
S35	τ(C6–C1–C2–C3) – τ(C1–C2–C3–N4) + τ(C2–C3–N4–C5) – τ(C3–N4–C5–C6) + τ(N4–C5–C6–C1) – τ(C5–C6–C1–C2)	τ_{Ring} 1
S36	τ(C6–C1–C2–C3) – τ(C2–C3–N4-C5) + τ(C3–N4–C5–C6) – τ(C5–C6–C1–C2)	τ_{Ring} 2
S37	τ(C6–C1–C2–C3) – 2τ(C1–C2–C3–N4) + τ(C2–C3–N4–C5) + τ(C3–N4–C5–C6) –2τ(N4–C5–C6–C1) + τ(C5–C6–C1–C2)	τ_{Ring} 3
S38	γ(H16–H17–N10–N9)	γ(NH_2)
S39	γ(N9–C1–C7–O8)	γ(C=O)
S40	γ(C2–C6–C1–C7)	γ(C–$CONHNH_2$)
S41	γ(N10–C7–N9–H15)	γ(N–H)
S42	γ(C1–C3–C2–H11) + γ(C2–N4–C3–H12) + γ(N4–C6–C5–H13) + γ(C1–C5–C6–H14)	γ(C–H) 1
S43	γ(C1–C3–C2–H11) – γ(C2–N4–C3–H12) + γ(N4–C6–C5–H13) – γ(C1–C5–C6–H14)	γ(C–H) 2
S44	γ(C1–C3–C2–H11) – γ(C2–N4–C3–H12) – γ(N4–C6–C5–H13) + γ(C1–C5–C6–H14)	γ(C–H) 3
S45	γ(C1–C3–C2–H11) + γ(C2–N4–C3–H12) – γ(N4–C6–C5–H13) – γ(C1–C5–C6–H14)	γ(C–H) 4

[a] *ν*, bond stretching; *δ*, bending; *γ*, rocking; *τ*, torsion; w, wagging; s, symmetric; as, asymmetric.

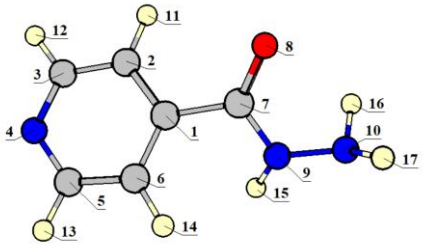

Table S11 – Definition of the internal coordinates used in the normal coordinate analysis of SUC.[a]

Coordinate	Definition	Approximate description
S1	ν(O4a–5a) + ν(O4b–H5b)	ν(O–H)s
S2	ν(O4a–5a) – ν(O4b–H5b)	ν(O–H)as
S3	ν(C2a–O4a) + ν(C2b–O4b)	ν(C–O)s
S4	ν(C2a–O4a) – ν(C2b–O4b)	ν(C–O)as
S5	ν(C2a–O3a) + ν(C2b–O3b)	ν(C=O)s
S6	ν(C2a–O3a) – ν(C2b–O3b)	ν(C=O)as
S7	ν(C1a–C2a) + ν(C1b–C2b)	ν(C–C)s
S8	ν(C1a–C2a) – ν(C1b–C2b)	ν(C–C)as
S9	ν(C1a–H7a) + ν(C1a–H6a) + ν(C1b–H7b) + ν(C1b–H6b)	ν(CH_2 s)s
S10	ν(C1a–H7a) + ν(C1a–H6a) – ν(C1b–H7b) – ν(C1b–H6b)	ν(CH_2 s)as
S11	ν(C1a–H7a) – ν(C1a–H6a) + ν(C1b–H7b) – ν(C1b–H6b)	ν(CH_2 as)s
S12	ν(C1a–H7a) – ν(C1a–H6a) – ν(C1b–H7b) + ν(C1b–H6b)	ν(CH_2 as)as
S13	ν(C1a–C1b)	ν(C–C)
S14	δ(H5a–C2a–O4a) + δ(H5b–C2b–O4b)	δ(COH)s
S15	δ(H5a–C2a–O4a) – δ(H5b–C2b–O4b)	δ(COH)as
S16	2δ(O4a–C1a–C2a) – δ(O4a–O3a–C2a) – δ(C1a–O3a–C2a) + 2δ(O4b–C1b–C2b) – δ(O4b–O3b–C2b) – δ(C1b–O3b–C2b)	δ(CCO)s
S17	2δ(O4a–C1a–C2a) – δ(O4a–O3a–C2a) – δ(C1a–O3a–C2a) – 2δ(O4b–C1b–C2b) +δ(O4b–O3b–C2b) + δ(C1b–O3b–C2b)	δ(CCO)as
S18	δ(O4a–O3a–C2a) – δ(C1a–O3a–C2a) + δ(O4b–O3b–C2b) – δ(C1b–O3b–C2b)	δ(C=O)s
S19	δ(O4a–O3a–C2a) – δ(C1a–O3a–C2a) – δ(O4b–O3b–C2b) + δ(C1b–O3b–C2b)	δ(C=O)as
S20	5δ(H7a–H6a–C1a) – δ(C1b–H6a–C1a) + δ(C1b–H7a–C1a) – δ(C2a–H7a–C1a) – δ(C2a–H6a–C1a) – δ(C1a–C2a–C1a) + 5δ(H7b–H6b–C1b) – δ(C1a–H6b–C1b) – δ(C1a–H7b–C1b) – δ(C2b–H7b–C1b) – δ(C2b–H6b–C1b) – δ(C1a–C2b–C1b)	δ(CH_2)s
S21	5δ(H7a–H6a–C1a) – δ(C1b–H6a–C1a) + δ(C1b–H7a–C1a) – δ(C2a–H7a–C1a) – δ(C2a–H6a–C1a) – δ(C1a–C2a–C1a) – 5δ(H7b–H6b–C1b) + δ(C1a–H6b–C1b) + δ(C1a–H7b–C1b) + δ(C2b–H7b–C1b) + δ(C2b–H6b–C1b) + δ(C1a–C2b–C1b)	δ(CH_2)as
S22	– δ(C1b–H6a–C1a) + δ(C1b–H7a–C1a) – δ(C2a–H7a–C1a) – δ(C2a–H6a–C1a) + 4δ(C1b–C2a–C1a) – δ(C1a–H6b–C1b) – δ(C1a–H7b–C1b) – δ(C1a–H6b–C1b) – δ(C2b–H7b–C1b) – δ(C2b–H6b–C1b) + 4δ(C1a–C2b–C1b)	δ(CCC)s
S23	– δ(C1b–H6a–C1a) + δ(C1b–H7a–C1a) – δ(C2a–H7a–C1a) – δ(C2a–H6a–C1a) + 4δ(C1b–C2a–C1a) + δ(C1a–H6b–C1b) + δ(C1a–H7b–C1b) + δ(C1a–H6b–C1b) + δ(C2b–H7b–C1b) + δ(C2b–H6b–C1b) – 4δ(C1a–C2b–C1b)	δ(CCC)as
S24	δ(C1b–H6a–C1a) + δ(C1b–H7a–C1a) – δ(C2a–H7a–C1a) – δ(C2a–H6a–C1a) + δ(C1a–H6b–C1b) + δ(C1a–H7b–C1b) – δ(C2b–H7b–C1b) – δ(C2b–H6b–C1b)	w(CH_2)s
S25	δ(C1b–H6a–C1a) + δ(C1b–H7a–C1a) – δ(C2a–H7a–C1a) – δ(C2a–H6a–C1a) – δ(C1a–H6b–C1b) – δ(C1a–H7b–C1b) + δ(C2b–H7b–C1b) + δ(C2b–H6b–C1b)	w(CH_2)as
S26	δ(C1b–H6a–C1a) – δ(C1b–H7a–C1a) + δ(C2a–H7a–C1a) – δ(C2a–H6a–C1a) + δ(C1a–H6b–C1b) – δ(C1a–H7b–C1b) + δ(C2b–H7b–C1b) – δ(C2b–H6b–C1b)	tw(CH_2)s
S27	δ(C1b–H6a–C1a) – δ(C1b–H7a–C1a) + δ(C2a–H7a–C1a) – δ(C2a–H6a–C1a) – δ(C1a–H6b–C1b) + δ(C1a–H7b–C1b) – δ(C2b–H7b–C1b) + δ(C2b–H6b–C1b)	tw(CH_2)as
S28	δ(C1b–H6a–C1a) – δ(C1b–H7a–C1a) – δ(C2a–H7a–C1a) + δ(C2a–H6a–C1a) + δ(C1a–H6b–C1b) – δ(C1a–H7b–C1b) – δ(C2b–H7b–C1b) + δ(C2b–H6b–C1b)	γ(CH_2)s
S29	δ(C1b–H6a–C1a) – δ(C1b–H7a–C1a) – δ(C2a–H7a–C1a) + δ(C2a–H6a–C1a) – δ(C1a–H6b–C1b) + δ(C1a–H7b–C1b) + δ(C2b–H7b–C1b) – δ(C2b–H6b–C1b)	γ(CH_2)as
S30	τ(O4a–C2a–C1a–C1b) + τ(O4b–C2b–C1b–C1a)	τ(C–C)s
S31	τ(O4a–C2a–C1a–C1b) – τ(O4b–C2b–C1b–C1a)	τ(C–C)as
S32	τ(C2a–C1a–C1b–C2b)	τ(C–C)
S33	τ(H5a–O4a–C2a–C1a) + τ(H5b–O4b–C2b–C1b)	τ(C–O)s
S34	τ(H5a–O4a–C2a–C1a) – τ(H5b–O4b–C2b–C1b)	τ(C–O)as
S35	γ(O4a–C1a–C2a–O3a) + γ(O4b–C1b–C2b–O3b)	γ(C=O)s
S36	γ(O4a–C1a–C2a–O3a) – γ(O4b–C1b–C2b–O3b)	γ(C=O)as

[a] *ν*, bond stretching; *δ*, bending; *γ*, rocking; *τ*, torsion; w, wagging; tw, twisting; s, symmetric; as, asymmetric.

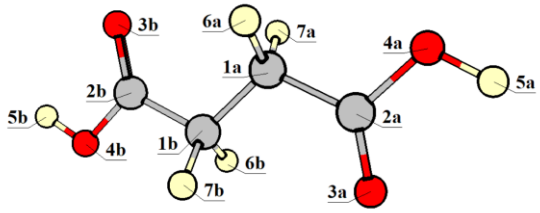

Table S12 – Definition of the internal coordinates used in the normal coordinate analysis of INH-SUC-INH trimer.[a]

Coordinate	Definition	Approximate description
S127	ν(H5b – N4') + ν(H13'–O3b)	ν(HB)s a
S128	ν(H5b – N4') – ν(H13'–O3b)	ν(HB)as a
S129	δ(N4'– O3b – H13') + δ(O3b – N4' – H5b) – δ(H5b – H13' – N4') – δ(H13'– H5b – O3b)	δ(im) 1 a
S130	τ(C5'– N4'– H13'– O3b) – τ(C5'– H13' – N4'– H5b) + τ(C2b– O3b – H5b–N4') – τ(O4b – H5b – O3b – H13')	τ(im) 1 a
S131	τ(C5'– N4'– H13'– O3b) – τ(C5'– H13' – N4'– H5b) – τ(C2b – O3b – H5b –N4') + τ(O4b – H5b – O3b – H13')	τ(im) 2 a
S132	τ(O3b– H13'– N4'– H5b) – τ(N4'– H5b – O3b – H13') – τ(H13'– N4' – H5b – O3b) + τ(H5b – O3b – H13' – N4')	τ(im) 3 a
S133	ν(H5a – N4) + ν(H13 – O3a)	ν(HB)s b
S134	ν(H5a – N4) – ν(H13 – O3a)	ν(HB)as b
S135	δ(N4 – O3a – H5a) + δ(N4 – O3a – H13) – δ(H13 – H5a – O3a) – δ(H5a – H13 – N4)	δ(im) 1 b
S136	τ(N4 – H5a – O3a – H13) + τ(O3a– H13 – N4 – H5a) – τ(H5a – O3a – H13 –N4) – τ(H13 – N4 – H5a – O3a)	τ(im) 1 b
S137	τ(C2a – O3a – H5a – N4) – τ(O4a– H5a – O3a – H13) + τ(C5 – N4 – H13 – O13) – τ(C5 – H13 – N4 – H5a)	τ(im) 2 b
S138	τ(C2a – O3a – H5a – N4) – τ(O4a– H5a – O3a – H13) – τ(C5 – N4 – H13 – O13) + τ(C5 – H13 – N4 – H5a)	τ(im) 3 b

[a] *ν*, bond stretching; *δ*, bending; *γ*, rocking; *τ*, torsion; s, symmetric; as, asymmetric.

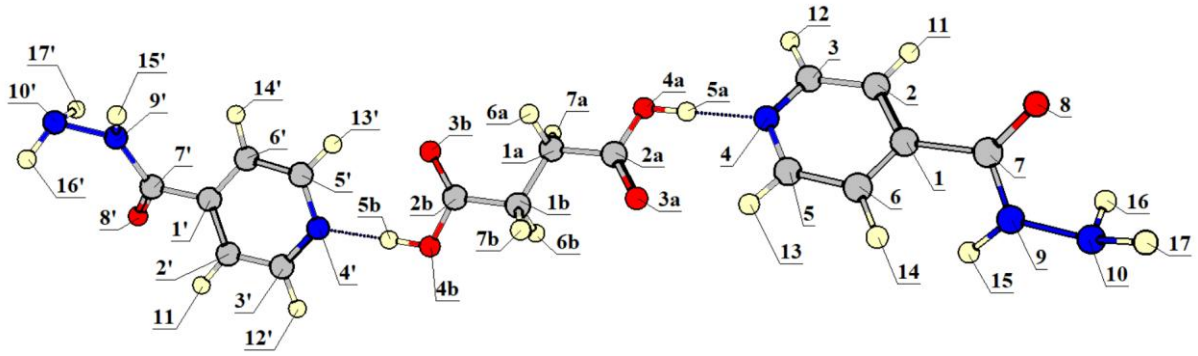

Table S13 – Definition of the internal coordinates used in the normal coordinate analysis of SUC-INH-INH-SUC tetramer.[a]

Coordinate	Definition	Approximate description
S163	ν(H13'– O3a') + ν(H5a'– N4')	ν(HB)s ca
S164	ν(H13'– O3a') – ν(H5a'– N4')	ν(HB)as ca
S165	δ(O3a'– N4' – H5a') + δ(N4' – O3a' – H13') – δ(H13' – H5a' – O3a') – δ(H5a'– H13' – N4')	δ(im) 1 ca
S166	τ(N4'– H5a'– O3a'– H13') – τ(O3a'– H13' – N4'– H5a') + τ(H5a'– O3a' – H13' – N4') – τ(H13' – N4' – H5a' – O3a')	τ(im) 1 ca
S167	τ(C2a'– O3a'– H5a'– N4') – τ(O4a'– H5a' – O3a'– H13') + τ(C5' – N4' – H13' – O3a') – τ(C5' – H13' – N4' – H5a')	τ(im) 2 ca
S168	τ(C2a'– O3a'– H5a'– N4') + τ(O4a'– H5a' – O3a'– H13') – τ(C5' – N4' – H13' – O3a') – τ(C5' – H13' – N4' – H5a')	τ(im) 3 ca
S169	ν(H17 – O8') + ν(H17' – O8)	ν(HB)s ab
S170	ν(H17 – O8') – ν(H17' – O8)	ν(HB)as ab
S171	δ(O8 – O8' – H17) + δ(O8' – O8 – H17') – δ(H17' – H17 – O8) – δ(H17 – H17' – O8')	δ(im) 1 ab
S172	τ(O8' – H17 – O8 – H17') + τ(O8– H17' – O8' – H17) – τ(H17 – O8 – H17' –O8') – τ(H17' – O8' – H17 – O8)	τ(im) 1 ab
S173	τ(C7 – O8 – H17 – O8') – τ(N10 – H17 – O8 – H17') + τ(C7' – O8' – H17' – O8) – τ(N10' – H17' – O8' – H17)	τ(im) 2 ab
S174	τ(C7 – O8 – H17 – O8') – τ(N10 – H17 – O8 – H17') – τ(C7' – O8' – H17' – O8) + τ(N10' – H17' – O8' – H17)	τ(im) 3 ab
S175	ν(H13' – O3a') + ν(H5a' – N4')	ν(HB)s bd
S176	ν(H13' – O3a') – ν(H5a' – N4')	ν(HB)as bd
S177	δ(O3a' – N4' – H5a') + δ(N4' – O3a' – H13') – δ(H13' – H5a' – O3a') – δ(H5a' – H13' – N4')	δ(im) 1 bd
S178	τ(N4' – H5a' – O3a' – H13') + τ(O3a'– H13' – N4' – H5a') – τ(H5a' – O3a' – H13' – N4') – τ(H13' – N4' – H5a' – O3a')	τ(im) 1 bd
S179	τ(C2a' – O3a' – H5a' – N4') – τ(O4a'– H5a' – O3a' – H13') + τ(C5' – N4' – H13' – O3a') – τ(C5' – H13' – N4' – H5a')	τ(im) 2 bd
S180	τ(C2a' – aO3a' – H5a' – N4') – τ(O4a'– H5a' – O3a' – H13') – τ(C5' – N4' – H13' – O3a') + τ(C5' – H13' – N4' – H5a')	τ(im) 3 bd

[a] *ν*, bond stretching; *δ*, bending; *γ*, rocking; *τ*, torsion; s, symmetric; as, asymmetric.

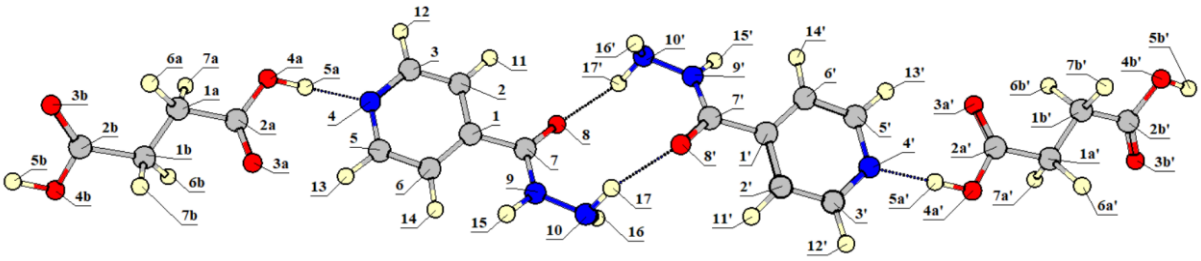

Table S14 – Definition of the internal coordinates used in the normal coordinate analysis of INH-INH dimer.[a]

Coordinate	Definition	Approximate description
S91	ν(H15' – N10) + ν(H15'– O10')	ν(HB)s
S92	ν(H15' – N10) + ν(H15'– O10')	ν(HB)as
S93	τ(N9 – N10 – H15 – N10') + τ(N9 – H15 – N10 – H15') – τ(N9' – N10' – H15' – N10) – τ(N9'– H15' – N10' – H15)	τ(im) 1
S94	δ(N10 – N10'– H15) – δ(N10'– N10 – H15') + δ(H15'– H15 – N10) – δ(H15 – H15' – N10)	δ(im)
S95	τ(N10'– H15– N10 – H15') + τ(N10 – H15' – N10'– H15) – τ(H15 – N10 – H15' –N10') – τ(H15' – N10' – H15 – N10)	τ(im) 2
S96	τ(N10'– H15– N10 – H15') – τ(N10 – H15' – N10'– H15) – τ(H15 – N10 – H15' –N10') + τ(H15' – N10' – H15 – N10)	τ(im) 3

[a] *ν*, bond stretching; *δ*, bending; *γ*, rocking; *τ*, torsion; s, symmetric; as, asymmetric.

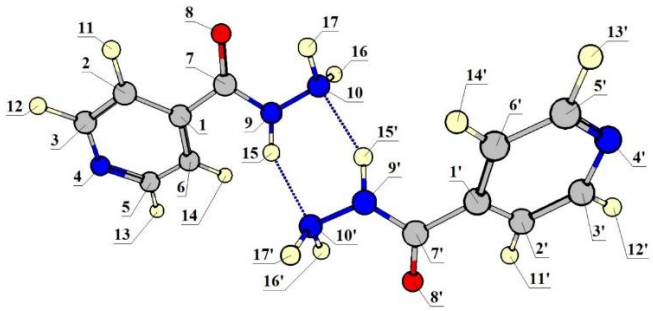

Table S15. Cartesian coordinates of the optimized geometry of INH molecule (Å).[a]

Atom	*x*	*y*	*z*
C1	-0.237495	0.103329	0.052717
C2	-1.087879	1.188319	-0.162804
C3	-2.459578	0.961546	-0.225425
N4	-3.019461	-0.244016	-0.075962
C5	-2.200845	-1.274566	0.148400
C6	-0.813537	-1.158278	0.219449
C7	1.243339	0.359991	0.124059
O8	1.701735	1.447204	0.437319
N9	2.045303	-0.707661	-0.186230
N10	3.443586	-0.567319	-0.279645
H11	-0.675516	2.183229	-0.273249
H12	-3.143069	1.786945	-0.401404
H13	-2.674509	-2.242646	0.283915
H14	-0.213494	-2.034741	0.434135
H15	1.670771	-1.475507	-0.722168
H16	3.641107	0.370570	-0.624892
H17	3.830808	-0.614546	0.659585

Table S16. Cartesian coordinates of the optimized geometry of SUC molecule (Å).[a]

Atom	*x*	*y*	*z*
C1a	0.557495	-0.519508	0.000047
C2a	1.929775	0.108585	-0.000002
O3a	2.172193	1.289862	-0.000004
O4a	2.902221	-0.835382	-0.000040
H5a	3.753398	-0.371966	-0.000067
H6a	0.483844	-1.180423	-0.868119
H7a	0.483889	-1.180367	0.868260
C1b	-0.557495	0.519508	0.000049
C2b	-1.929775	-0.108585	-0.000001
O3b	-2.172193	-1.289862	-0.000003
O4b	-2.902221	0.835382	-0.000042
H5b	-3.753398	0.371966	-0.000068
H6b	-0.483890	1.180365	0.868265
H7b	-0.483844	1.180424	-0.868115

Table S17. Cartesian coordinates of the optimized geometry of INH-SUC-INH trimer molecule (Å).[a]

Atom	*x*	*y*	*z*
C1	-7.954300	-0.304796	0.158461
C2	-7.629776	-1.561818	-0.354822
C3	-6.294612	-1.862687	-0.592270
N4	-5.297919	-1.004774	-0.338727
C5	-5.603966	0.194175	0.170683
C6	-6.914385	0.585597	0.433464
C7	-9.406105	0.002072	0.417517
O8	-10.226458	-0.877739	0.623764
N9	-9.742284	1.329149	0.405149
N10	-11.082399	1.746297	0.515127
H11	-8.412603	-2.281891	-0.554960
H12	-6.006980	-2.827012	-0.998596
H13	-4.758514	0.844677	0.371245
H14	-7.098289	1.558056	0.873857
H15	-9.135300	1.999002	-0.042132
H16	-11.666295	1.050263	0.054555
H17	-11.340166	1.733651	1.498858
C1a	-0.405676	-0.592060	-0.549784
C2a	-1.894287	-0.381301	-0.375828
O3a	-2.398058	0.632083	0.069797
O4a	-2.598188	-1.450531	-0.752416
H5a	-3.575528	-1.288901	-0.599814
H6a	-0.088905	-1.336836	0.187059
H7a	-0.231923	-1.059326	-1.522323
C1b	0.402692	0.689543	-0.389115
C2b	1.894607	0.445481	-0.316236
O3b	2.406090	-0.647712	-0.166007
O4b	2.592431	1.578392	-0.417077
H5b	3.572773	1.385400	-0.337757
H6b	0.205087	1.395470	-1.199668
H7b	0.108182	1.213228	0.525764
C1’	7.964225	0.250479	0.045220
C2’	7.633289	1.595047	-0.132276
C3’	6.293517	1.944894	-0.242665
N4’	5.300013	1.047775	-0.199705
C5’	5.614750	-0.243969	-0.048076
C6’	6.929202	-0.686231	0.078772
C7’	9.422059	-0.112496	0.154832
O8’	10.300496	0.586319	-0.324347
N9’	9.694271	-1.274124	0.826234
N10’	11.017511	-1.667521	1.102531
H11’	8.415535	2.341046	-0.186579
H12’	5.999736	2.981451	-0.373050
H13’	4.773643	-0.929911	-0.034571
H14’	7.124180	-1.747946	0.169219
H15’	9.005947	-1.670453	1.447792
H16’	11.568597	-0.825463	1.259909
H17’	11.398578	-2.110196	0.269968

Table S18. Cartesian coordinates of the optimized geometry of SUC-INH-INH-SUC tetramer molecule (Å).[a]

Atom	*x*	*y*	*z*
C1	-3.849309	-0.064659	0.397460
C2	-4.170396	-1.422603	0.348608
C3	-5.505552	-1.792833	0.251599
N4	-6.507133	-0.904417	0.217516
C5	-6.205402	0.397393	0.284625
C6	-4.895202	0.860505	0.373947
C7	-2.394445	0.315396	0.508251
O8	-1.575092	-0.449179	1.007342
N9	-2.063987	1.539141	0.023994
N10	-0.766601	2.076182	0.072024
H11	-3.385285	-2.166000	0.393611
H12	-5.789008	-2.839192	0.202541
H13	-7.053599	1.074402	0.269492
H14	-4.719622	1.926345	0.455966
H15	-2.741166	2.104152	-0.462917
H16	-0.473990	2.110979	1.045150
H17	-0.118416	1.457524	-0.418207
C1a	-11.393912	-0.643639	-0.109076
C2a	-9.910736	-0.367942	0.024555
O3a	-9.425683	0.745193	0.093853
O4a	-9.194555	-1.490954	0.040918
H5a	-8.215859	-1.275498	0.105510
H6a	-11.564873	-1.070003	-1.102239
H7a	-11.665987	-1.434573	0.594214
C1b	-12.245310	0.604090	0.092608
C2b	-13.698439	0.382278	-0.245669
O3b	-14.168181	-0.590381	-0.782397
O4b	-14.458304	1.446267	0.118966
H5b	-15.369980	1.248953	-0.143713
H6b	-12.180361	0.975446	1.118657
H7b	-11.878980	1.425042	-0.531545
C1’	3.849340	0.065042	-0.397763
C2’	4.170445	1.422981	-0.348892
C3’	5.505605	1.793195	-0.251888
N4’	6.507180	0.904772	-0.217822
C5’	6.205429	-0.397038	-0.284957
C6’	4.895223	-0.860132	-0.374276
C7’	2.394471	-0.314992	-0.508564
O8’	1.575126	0.449618	-1.007614
N9’	2.063990	-1.538732	-0.024309
N10’	0.766593	-2.075747	-0.072331
H11’	3.385343	2.166389	-0.393881
H12’	5.789071	2.839550	-0.202815
H13’	7.053618	-1.074059	-0.269878
H14’	4.719635	-1.925968	-0.456322
H15’	2.741167	-2.103770	0.462575
H16’	0.473984	-2.110553	-1.045458

H17’	0.118424	-1.457065	0.417887
C1a’	11.393944	0.643155	0.109029
C2a’	9.910715	0.367652	-0.024379
O3a’	9.425461	-0.745426	-0.093185
O4a’	9.194742	1.490788	-0.041269
H5a’	8.216005	1.275529	-0.105898
H6a’	11.666275	1.432995	-0.595395
H7a’	11.564933	1.070965	1.101561
C1b’	12.245043	-0.605066	-0.090837
C2b’	13.698438	-0.382764	0.245969
O3b’	14.168856	0.591375	0.779409
O4b’	14.457708	-1.448132	-0.115886
H5b’	15.369630	-1.250299	0.145540
H6b’	11.879051	-1.424856	0.535026
H7b’	12.179374	-0.978300	-1.116165

Table S19. Cartesian coordinates of the optimized geometry of INH-INH dimer molecule (Å).[a]

Atom	*x*	*y*	*z*
C1	0.331526	3.750613	-0.210240
C2	0.788250	4.931348	0.376538
C3	-0.097265	5.996171	0.514897
N4	-1.369783	5.959923	0.104840
C5	-1.795102	4.831802	-0.469509
C6	-0.993817	3.705709	-0.649053
C7	1.298479	2.609090	-0.357890
O8	2.510374	2.772058	-0.317080
N9	0.738494	1.373656	-0.541351
N10	1.579838	0.232084	-0.542955
H11	1.815657	5.004792	0.709697
H12	0.230146	6.923903	0.974840
H13	-2.827023	4.826409	-0.808668
H14	-1.400509	2.831868	-1.143302
H15	-0.206211	1.164173	-0.217790
H16	1.875375	0.060918	-1.502666
H17	2.429500	0.489073	-0.037927
C1’	-0.331526	-3.750613	0.210240
C2’	-0.788250	-4.931348	-0.376538
C3’	0.097265	-5.996171	-0.514897
N4’	1.369783	-5.959923	-0.104840
C5’	1.795102	-4.831802	0.469509
C6’	0.993817	-3.705709	0.649053
C7’	-1.298479	-2.609090	0.357890
O8’	-2.510374	-2.772058	0.317080
N9’	-0.738494	-1.373656	0.541351
N10’	-1.579838	-0.232084	0.542955
H11’	-1.815657	-5.004792	-0.709697
H12’	-0.230146	-6.923903	-0.974840
H13’	2.827023	-4.826409	0.808668
H14’	1.400509	-2.831868	1.143302
H15’	0.206211	-1.164173	0.217790
H16’	-2.429500	-0.489073	0.037927
H17’	-1.875375	-0.060918	1.502666

Table S20. Assignment of the IR and Raman spectra for INH crystal (polymorph 1).[a]

ṽ (IR)	ṽ (Raman)	ṽ (calc)[b]	I_{IR}[b]	ṽ (calc)[c]	I_{IR}[c]	a_R[c]	PED (%)[d]
3302	3304	3462	32	3620	43	53	99% ν(N–H)
3243	3270/3246	3351	2	3529	9	62	99% ν(NH_2)as
3210	3204	3317	0.5	3462	2	149	98% ν(NH_2)s
3103	3109	3072	4	3204	2	93	97% ν(C–H) 3
	3097	3061	23	3184	7	108	93% ν(C–H) 4
3047	3061	3025	13	3157	12	121	89% ν(C–H) 1
		3018	7	3152	20	107	86% ν(C–H) 2
1661	1673	1701	159	1735	197	45	79% ν(C=O)
1631	1646	1641	27	1707	72	5	94% δ(NH_2)
1600	1605	1589	9	1632	12	43	64% ν_{Ring} 3, 22% δ(C–H) 2
1552	1556	1555	22	1597	33	4	74% ν_{Ring} 2
1491	1497	1491	4	1524	34	7	52% δ(C–H) 4, 26% ν_{Ring} 6
1463		1468	15	1498	158	7	57% δ(N–H), 15% ν(N–C), 11% δ(C–H) 4
1409	1415	1413	4	1437	12	1	50% δ(C–H) 3, 32% ν_{Ring} 5
1331	1335	1336	2	1351	2	1	55% δ(C–H) 1, 28% w(NH_2)
1323		1301	2	1350	1	7	69% w(NH_2), 22% δ(C–H) 1
		1267	84	1303	125	32	21% ν(N–C), 19% δ(N–H), 16% ν(C–C), 11% ν(N–N)
1263	1224	1237	1	1276	9	6	75% ν_{Ring} 4
1220/1194	1190	1219	1	1244	2	5	58% δ(C–H) 2, 25% ν_{Ring} 3
1139	1132	1175	3	1210	3	34	28% ν(N–N), 18% γ(NH_2), 17% ν(C–C), 12% ν_{Ring} 1, 10% δ
1097	1096	1096	1	1116	2	2	22% ν_{Ring} 5, 20% δ(C–H) 3, 15% ν(N–C), 12% γ(NH_2)
1060	1060	1076	1	1103	1	2	23% ν_{Ring} 5, 20% δ(C–H) 3, 13% ν(N–C), 12% ν_{Ring} 6
1046		1067	7	1089	6	2	27% ν_{Ring} 6, 24% δ_{Ring} 1, 15% δ(C–H) 4
965	1002	996	1	1011	3	32	53% ν_{Ring} 1, 42% δ_{Ring} 1
		996	1	1006	1	0.4	51% γ(C–H) 2, 26% γ(C–H) 3, 16% γ(C–H) 4
945	964	975	5	984	2	1	42% γ(C–H) 4, 31% γ(C–H) 1, 16% γ(C–H) 3
1004/993	942	920	87	955	89	3	42% ν(N–N), 33% γ(NH_2)
886	889	885	2	895	3	0.4	45% γ(C–H) 1, 21% γ(C–H) 3, 15% γ(C–H) 4
877		858	2	892	9	9	17% δ(C=O), 15% γ(NH_2), 14% δ(CNN)
844	850	844	12	855	27	1	27% γ(C–H) 3, 19% γ(C–H) 2, 19% γ(C=O), 15% γ(C–$CONHNH_2$)
740	756/728	762	5	771	14	0.3	46% τ_{Ring} 1, 28% γ(C=O)
671	686	719	14	724	18	1	60% τ_{Ring} 1, 35% γ(C=O)
656		686	13	691	17	3	55% δ_{Ring} 3, 14% δ(C=O)
n.obs.	668	675	0.3	681	1	7	88% δ_{Ring} 2
505	508	549	13	525	71	1	30% γ(N–H), 20% τ_{Ring} 2, 13% γ(C–$CONHNH_2$)
437	439	436	55	523	63	3	51% γ(N–H), 13% δ(CCO)
n.i.	403	410	31	424	10	1	34% τ_{Ring} 2, 17% δ(C=O)
n.i.	n.obs.	380	0.1	385	0.1	0.1	98% τ_{Ring} 3
n.i.	367	343	12	347	6	6	34% ν(C–C), 28% δ_{Ring} 3, 11% δ(CNN)
n.i.	329	302	27	304	35	1	32% δ(CNN), 20% w(C–$CONHNH_2$), 19% δ(C=O)
n.i.	256/225	232	22	239	10	1	49% τ(C–N), 23% τ_{Ring} 2
n.i.	176	159	4	154	8	2	39% τ(N–N), 30% w(C–$CONHNH_2$), 20% δ(CCO)
n.i.	164	127	7	136	9	2	50% τ(N–N), 15% γ(N–H), 11% δ(CCO)
n.i.	125	103	2	113	4	1	41% τ(C–N), 36% γ(C–$CONHNH_2$), 13% τ_{Ring} 2
n.i.	n.i.	35	2	55	1	2	95% τ(C–C)

[a] Wavenumbers (ṽ) in cm^{-1}; calculated infrared intensities (I_{IR}) in km mol^{-1}; calculated Raman scattering activities (a_R) in $Å^4$ u^{-1}; ν, bond stretching; δ, bending; γ, rocking; τ, torsion; w, wagging; s, symmetric; as, anti-symmetric; n.obs., non-observed; n.i., non-investigated. [b] Anharmonic wavenumbers and IR intensities. [c] Harmonic wavenumbers, IR intensities and Raman activities. [d] For definition of coordinates see Table S10; only PED values ≥ 10% are shown.

Table S21. Assignment of the IR and Raman spectra for SUC crystal (polymorph β).[a]

ϋ (IR)	ϋ (Raman)	ϋ (calc)[b]	I_{IR}[b]	ϋ (calc)[c]	I_{IR}[c]	a_R[c]	PED (%)[d]
3380-2170		3580	0	3758	0	257	100% ν(O–H)s
	3150-2750	3580	119	3758	144	0	100% ν(O–H)as
2981		2953	9	3097	4	0	100% ν(CH_2 as)as
	2966	2937	0	3078	0	90	100% ν(CH_2 as)s
2930		2944	11	3062	10	0	100% ν(CH_2 s)as
	2925	2933	0	3055	0	199	100% ν(CH_2 s)s
	1652	1780	0	1813	0	19	84% ν(C=O)s
1675		1780	439	1807	625	0	86% ν(C=O)as
1410		1430	26	1464	46	0	97% δ(CH)as
	1464	1430	0	1463	0	10	79% δ(CH_2)s, 14% w(CH_2)s
	1369	1387	0	1430	0	2	49% w(CH_2)s, 18% δ(CH_2)s, 12% ν(C–C)s
1301		1340	72	1377	203	0	29% w(CH_2)as, 18% δ(COH)as, 17% ν(C–O)as, 16% ν(C–C)as, 15% δ(C=O)as
	1289	1224	0	1312	0	7	51% δ(COH)s, 15% w(CH_2)s, 12% δ(C=O)s, 10% ν(C–O)s
	1225	1273	0	1310	0	8	93% tw(CH_2)s
1194		1217	1	1275	35	0	45% δ(COH)as, 44% w(CH_2)as
1175		1166	0.03	1189	1	0	85% tw(CH_2)as, 10% γ(C=O)s
	1080	1194	0	1165	0	4	43% ν(C–O)s, 24% δ(COH)s
905/888		1109	578	1132	571	0	47% ν(C–O)as, 22% w(CH_2)as, 21% δ(COH)as
	1028	988	0	1080	0	1	67% ν(C–C), 14% ν(C–O)s
	995	1020	0	1044	0	1	69% γ(CH_2)s, 28% γ(C=O)as
	933	908	0	917	0	16	49% ν(C–C)s, 24% δ(CCC)s
888		850	5	868	4	0	61% ν(C–C)as, 26% ν(C–O)as
801		805	16	809	33	0	47% γ(CH_2)as, 39% γ(C=O)s, 14% tw(CH_2)as
	682	650	0	655	0	9	61% δ(C=O)s, 19% ν(C–O)s
		649	0	653	0	1	55% τ(C–O)as, 35% γ(C=O)as
636		635	143	643	182	0	63% τ(C–O)s, 19% γ(CH_2)as, 18% γ(C=O)s
581		600	54	599	66	0	77% δ(C=O)as, 11% ν(C–C)as
545		516	61	531	55	0	74% δ(CCO)as, 17% δ(CCC)as
	580	489	0	516	0	4	46% τ(C–O)as, 34% γ(C=O)as, 14% γ(CH_2)s
443/417		524	77	514	55	0	37% τ(C–O)s, 30% γ(CH_2)as, 29% γ(C=O)s
	396	360	0	359	0	2	70% δ(CCO)s, 15% ν(C–C), 11% ν(C–C)s
	312	255	0	248	0	1	51% δ(CCC)s, 29% δ(CCO)s, 17% ν(C–C)s
n.i.		135	3	129	5	0	74% δ(CCC)as, 23% δ(CCO)as
	156	118	0	78	0	1	94% τ(C–C)as
n.i.	n.i.	96	0.2	69	0.2	0	68% τ(C–C), 32% τ(C–C)s
n.i.	n.i.	3	4	3	4	0	67% τ(C–C)s, 30% τ(C–C)

[a] Wavenumbers (ϋ) in cm^{-1}; calculated infrared intensities (I_{IR}) in km mol^{-1}; calculated Raman scattering activities (a_R) in $Å^4$ u^{-1}; ν, bond stretching; δ, bending; γ, rocking; τ, torsion; w, wagging; tw, twisting; s, symmetric; as, anti-symmetric; n.obs., non-observed; n.i., non-investigated. [b] Anharmonic wavenumbers and IR intensities. [c] Harmonic wavenumbers, IR intensities and Raman activities. [c] For definition of coordinates see Table S11; only PED values ≥ 10% are shown.

Table S22. Assignment of the IR and Raman spectra for INH-SUC cocrystal.[a]

INH-SUC cocrystal; Exp.		INH-SUC cocrystal; Fully Periodic B3LYP/6-31G(d,p)-D3									
		A_u		B_u		A_g		B_g			
ϑ̃ (IR)	ϑ̃ (Raman)	ϑ̃	I_{IR}	ϑ̃	I_{IR}	ϑ̃	a_R[b]	ϑ̃	a_R[b]	DS[c]	Assignment[b]
3300	3306	3311	77	3311	951	3315	122	3315	67	4	ν(NH_2)as
3249	3281	3251	773	3253	404	3250	436	3250	150	3	ν(NH_2)s
3218	3209	3150	668	3153	4535	3145	562	3144	56	9	ν(N–H)
3163	3147	3104	24	3104	76	3104	540	3104	35	0	ν(C–H)
3098	3091	3097	166	3098	137	3098	78	3096	78	2	ν(C–H)
3076	3073	3078	0	3079	7	3078	264	3078	12	1	ν(C–H)
3045/3025	3047	3066	5	3066	0	3066	103	3066	67	0	ν(C–H)
2977	2999	2994	1	2994	31	2980	123	2980	106	14	ν(CH_2)as
2937	2932	2939	8	2939	5	2931	379	2929	77	10	ν(CH_2)s
2402/1977	2473	2144	19078	2191	5771	2200	729	2238	328	94	ν(O–H)
1707	1702	1706	103	1705	1691	1709	146	1710	0	5	ν(C=O) INH
1675	1661	1683	435	1685	18	1679	8	1680	8	6	ν(C=O) SUC
1630	1636	1661	107	1660	3200	1651	416	1654	162	10	δ(NH_2)
1609	1608	1620	72	1619	111	1619	226	1619	79	1	ν_{Ring}
1536	1559	1575	85	1578	25	1579	57	1580	19	5	ν_{Ring}
		1559	2303	1570	429	1568	49	1567	2	11	δ(N–H)
	1540	1523	53	1522	208	1525	7	1527	2	5	δ(COH)
1495	1497	1501	0	1497	0	1496	29	1495	5	6	δ(C–H)
1424	1420	1442	139	1442	21	1443	60	1443	29	1	δ(CH_2)
1407	1389	1410	66	1410	419	1411	14	1417	4	7	δ(C–H)
1319	1339	1354	426	1356	961	1400	52	1402	17	48	w(NH_2), ν(C–O)
		1344	748	1343	965	1357	537	1359	4	16	δ(N–H)
		1324	0	1326	62	1325	9	1324	9	2	δ(C–H)
1288	1278	1288	49	1288	53	1285	7	1284	60	4	γ(NH_2), w(CH_2)
		1277	4	1278	100	1279	25	1282	31	5	ν_{Ring}
1233	1225	1209	21	1212	56	1276	97	1276	3	67	δ(C–H)
1212	1213	1192	35	1198	207	1257	43	1258	28	66	w(CH_2)
1185	1183	1188	34	1183	473	1211	96	1208	41	28	w(CH_2)
1167	1127	1158	6	1158	19	1198	510	1196	1	40	tw(CH_2)
1140		1152	54	1153	521	1156	7	1155	6	4	τ(C–O)
1104	1098	1123	179	1126	20	1124	20	1127	17	4	w(NH_2), ν(N–C)
1088	1084	1100	2	1100	136	1100	52	1100	13	0	δ(C–H)
1052	1076	1062	183	1061	126	1074	18	1075	21	14	δ(C–H), ν_{Ring}
	1056	1021	97	1022	37	1061	97	1065	0	44	δ_{Ring}
989	1018	997	291	1000	21	1020	118	1022	2	25	ν(N–N), γ(NH_2)
		992	40	982	81	1019	182	1020	1	38	γ(C–H)
968	971	962	4	974	12	1003	15	1006	3	44	γ(C–H)
960	929	896	0	896	493	983	4	992	0	96	γ(N–H)
885	880	891	186	894	5	975	56	962	7	84	ν(C–C), γ(N–H)
875		877	17	880	77	925	91	925	4	48	ν(N–C), δ(CNN)

849	848	868	22	871	216	893	349	888	7	25	γ(C–H)
800		834	6	830	40	879	37	874	5	49	γ(C–H)
783	795	789	16	788	88	846	23	851	15	63	γ(C=O) SUC
739	772	744	36	745	262	799	18	800	9	56	τ_{Ring}
693	694	701	192	697	53	745	16	745	9	48	δ_{Ring}
673		681	120	679	480	701	65	704	11	25	γ(C=O) INH
661	633	664	6	667	70	697	4	698	34	34	δ_{Ring}
635		630	70	631	13	682	13	684	15	54	δ(C=O) SUC
542	571	555	24	555	3	665	104	660	68	110	γ(CH_2)
		547	96	549	14	566	5	565	9	19	δ(CCO)
521	501	514	4	506	10	556	19	557	22	51	δ(CCC)
485		497	68	499	71	503	8	510	18	13	τ(N–N)
437	434	437	43	434	56	442	51	440	67	8	τ_{Ring}
n.i.	406	400	4	387	194	425	48	433	14	46	τ_{Ring}
n.i.	391	381	84	384	66	386	6	398	0	17	δ(CNN)
n.i.	364	355	1	357	4	369	69	364	10	14	δ(CNN)
n.i.	254	259	60	262	50	352	5	353	46	94	w(C–$CONHNH_2$)
n.i.	225	242	118	253	117	274	78	269	0	32	δ(CCC)
n.i.		232	27	229	143	265	36	263	14	36	δ(CCC)
n.i.	163	195	0	194	12	249	40	247	25	55	Skeletal
n.i.		191	9	191	11	214	117	211	95	23	Skeletal
n.i.		159	55	179	119	203	53	192	10	44	Skeletal
n.i.		154	29	153	2	165	190	162	287	12	Intermolecular
n.i.	n.i.	135	3	134	5	154	102	157	33	23	τ(C–N)
n.i.	n.i.	119	4	117	17	145	1000	146	87	29	Intermolecular
n.i.	n.i.	112	5	108	0	139	212	135	71	31	γ(C–$CONHNH_2$)
n.i.	n.i.	93	2	88	28	132	745	128	178	44	τ(C–C) INH
n.i.	n.i.	83	0	74	2	125	135	111	324	51	τ(C–C) SUC
n.i.	n.i.	52	1	59	4	100	346	101	92	49	τ(C–C) SUC
n.i.	n.i.	46	1	36	1	85	211	91	472	55	Intermolecular
n.i.	n.i.	13	0			68	45	73	419	60	Intermolecular
n.i.	n.i.					27	60	62	498	35	Intermolecular

[a] Wavenumbers (ῡ) in cm^{-1}; calculated infrared intensities (I_{IR}) in km mol^{-1}; calculated Raman scattering activities (a_R) in $Å^4$ u^{-1}; ν, bond stretching; δ, bending; γ, rocking; τ, torsion; w, wagging; tw, twisting; s, symmetric; as, anti-symmetric; n.i., non-investigated; calculated wavenumbers were scaled as described in the Materials, and Experimental and Computational Methods section of this article. [b] Raman activities are normalized so that the activity of the highest band is 1000. [c] Davidov splitting (DS) for each mode was calculated as the difference between the highest and smallest wavenumber among the four related modes of different symmetry. [b] Based on the vibration's animation mode of Chemcraft (G. A. Zhurko, Chemcraft - graphical program for visualization of quantum chemistry computations. Ivanovo, Russia, 2005. https://chemcraftprog.com).

Table S23. Results of the normal coordinate analysis for INH-SUC-INH.[a]

Nº	ῡ	PED (≥10%)
138	3620	99% ν(N–H)
137	3620	99% ν(N–H)
136	3531	99% ν(NH_2)as
135	3531	99% ν(NH_2)as
134	3464	99% ν(NH_2)s
133	3464	99% ν(NH_2)s
132	3209	96% ν(C–H) 3
131	3209	96% ν(C–H) 3
130	3191	85% ν(C–H) 4, 14% ν(C–H) 2
129	3191	85% ν(C–H) 4, 14% ν(C–H) 2
128	3173	82% ν(C–H) 2, 15% ν(C–H) 4
127	3173	81% ν(C–H) 2, 14% ν(C–H) 4
126	3168	92% ν(C–H) 1
125	3168	92% ν(C–H) 1
124	3097	99% ν(O–H)s
123	3095	97% ν(CH_2 as)as
122	3086	95% ν(O–H)as
121	3076	91% ν(CH_2 as)s
120	3055	89% ν(CH_2 s)as
119	3050	97% ν(CH_2 s)s
118	1763	79% ν(C=O)s
117	1759	81% ν(C=O)as
116	1735	80% ν(C=O)
115	1735	80% ν(C=O)
114	1706	95% δ(NH_2)
113	1706	95% δ(NH_2)
112	1643	65% ν_{Ring} 3, 23% δ(C–H) 2
111	1643	65% ν_{Ring} 3, 23% δ(C–H) 2
110	1597	74% ν_{Ring} 2
109	1596	74% ν_{Ring} 2
107	1528	53% δ(C–H) 4, 27% ν_{Ring} 6
108	1528	53% δ(C–H) 4, 27% ν_{Ring} 6
106	1501	56% δ(N–H), 15% ν(N–C), 11% δ(C–H) 4
105	1501	57% δ(N–H), 15% ν(N–C), 11% δ(C–H) 4
104	1485	51% δ(COH)s
103	1483	51% δ(COH)as
102	1466	87% δ(CH)as
101	1464	89% δ(CH_2)s
100	1443	44% δ(C–H) 3, 31% ν_{Ring} 5
99	1442	43% δ(C–H) 3, 31% ν_{Ring} 5
98	1407	49% w(CH_2)s, 16% δ(COH)s
97	1354	41% δ(C–H) 1, 15% w(CH_2)as
96	1352	75% δ(C–H) 1, 11% ν_{Ring} 2
95	1350	91% w(NH_2)
94	1350	89% w(NH_2)
93	1349	37% δ(C–H) 1, 18% ω(CH_2)as
92	1304	21% ν(N–C), 20% δ(N–H), 15% ν(C–C), 11% ν(N–N)
91	1304	21% ν(N–C), 19% δ(N–H), 15% ν(C–C), 11% ν(N–N)
90	1297	74% tw(CH_2)s
89	1280	74% ν_{Ring} 4
88	1280	75% ν_{Ring} 4
87	1251	54% ν(C–O)s, 17% w(CH_2)s, 11% δ(C=O)s

86	1245	57% δ(C–H) 2, 25% ν_{Ring} 3
85	1245	58% δ(C–H) 2, 25% ν_{Ring} 3
84	1210	29% ν(N–N), 17% ν(C–C), 15% γ(NH_2), 11% ν_{Ring} 1, 10% δ_{Ring} 1
83	1210	29% ν(N–N), 17% ν(C–C), 15% γ(NH_2), 11% ν_{Ring} 1, 10% δ_{Ring} 1
82	1199	49% w(CH_2)as, 38% ν(C–O)as
81	1184	83% tw(CH_2)as
80	1118	27% ν_{Ring} 5, 26% δ(C–H) 3, 10% ν(N–C)
79	1118	27% ν_{Ring} 5, 26% δ(C–H) 3, 10% ν(N–C)
78	1106	18% ν(N–C), 18% ν_{Ring} 5, 17% δ(C–H) 3, 13% γ(NH_2)
77	1106	18% ν_{Ring} 5, 18% ν(N–C), 17% δ(C–H) 3, 13% γ(NH_2)
76	1088	37% ν(C–C), 16% ν_{Ring} 6, 11% δ_{Ring} 1
75	1088	34% ν(C–C), 16% ν_{Ring} 6, 11% δ_{Ring} 1
74	1088	32% ν_{Ring} 6, 22% δ_{Ring} 1, 15% δ(C–H) 4, 10% ν_{Ring} 1
73	1044	68% γ(CH_2)s, 27% γ(C=O)as
72	1027	45% ν_{Ring} 1, 42% δ_{Ring} 1
71	1027	45% ν_{Ring} 1, 42% δ_{Ring} 1
70	1025	35% γ(C–H) 4, 24% γ(C–H) 1, 16% τ(im) 2 a, 11% γ(C–H) 2
69	1024	33% γ(C–H) 4, 23% γ(C–H) 1, 17% τ(im) 3 b, 10% γ(C–H) 2
68	1017	31% τ(C–O)s, 11% τ(im) 1 b, 11% γ(C–H) 4, 11% τ(im) 3 b
67	1016	31% τ(C–O)as, 12% τ(im) 2 a, 11% τ(im) 3 a, 10% γ(C–H) 4
66	993	42% γ(C–H) 3, 39% γ(C–H) 2
65	993	42% γ(C–H) 3, 39% γ(C–H) 2
64	954	41% ν(N–N), 34% γ(NH_2)
63	954	41% ν(N–N), 34% γ(NH_2)
62	937	44% ν(C–C)s, 21% δ(CCC)s
61	898	37% γ(C–H) 1, 27% γ(C–H) 3, 14% γ(C–H) 2
60	898	37% γ(C–H) 1, 27% γ(C–H) 3, 14% γ(C–H) 2
59	895	55% ν(C–C)as, 25% ν(C–O)as
58	892	15% δ(C=O), 12% δ(CNN), 12% γ(NH_2), 12% γ(C–H) 1
57	892	15% δ(C=O), 12% δ(CNN), 12% γ(NH_2), 12% γ(C–H) 1
56	866	23% γ(C–H) 4, 16% γ(C–H) 3, 15% γ(C=O), 15% γ(C–$CONHNH_2$), 12% γ(C–H) 2
55	866	23% γ(C–H) 4, 16% γ(C–H) 3, 15% γ(C=O), 15% γ(C–$CONHNH_2$), 12% γ(C–H) 2
54	810	46% γ(CH_2)as, 33% γ(C=O)s, 14% tw(CH_2)as
53	775	41% τ_{Ring} 1, 33% γ(C=O)
52	775	41% τ_{Ring} 1, 33% γ(C=O)
51	722	61% τ_{Ring} 1, 32% γ(C=O)
50	722	62% τ_{Ring} 1, 32% γ(C=O)
49	703	54% δ_{Ring} 3, 13% δ(C=O)
48	703	54% δ_{Ring} 3, 13% δ(C=O)
47	696	58% δ(C=O)s, 15% ν(C–O)s, 12% δ(CCC)s
46	679	87% δ_{Ring} 2
45	678	84% δ_{Ring} 2
44	637	67% δ(C=O)as, 17% ν(C–C)as
43	601	44% γ(C=O)as, 22% δ(C=O)as, 11% γ(CH_2)s
42	564	47% γ(C=O)s, 44% γ(CH_2)as
41	532	51% δ(C=O)as, 16% γ(C=O)as
40	526	26% τ_{Ring} 2, 17% γ(C–$CONHNH_2$), 10% δ(CCO)
39	526	24% τ_{Ring} 2, 16% γ(C–$CONHNH_2$), 12% γ(N–H)
38	522	70% γ(N–H)
37	521	71% γ(N–H)
36	426	33% τ_{Ring} 2, 17% δ(C=O)
35	425	34% τ_{Ring} 2, 17% δ(C=O)
34	404	51% δ(C=O)s, 13% ν(C–C)
33	396	96% τ_{Ring} 3

32	396	96% τ_{Ring} 3
31	357	33% ν(C–C), 22% δ_{Ring} 3
30	353	32% ν(C–C), 23% δ_{Ring} 3
29	305	34% δ(CNN), 20% w(C–CONHNH$_2$), 19% δ(C=O)
28	305	34% δ(CNN), 20% w(C–CONHNH$_2$), 19% δ(C=O)
27	270	46% δ(CCC)s, 21% δ(C=O)s, 16% ν(C–C)s
26	246	43% τ(C–N), 21% τ_{Ring} 2
25	245	43% τ(C–N), 21% τ_{Ring} 2
24	204	31% δ(CCC)as, 14% δ(im) 1 a, 14% δ(im) 1 b
23	166	22% δ(CCO), 21% w(C–CONHNH$_2$)
22	160	25% δ(CCO), 25% w(C–CONHNH$_2$), 13% τ(N–N)
21	141	44% τ(N–N), 15% γ(C–CONHNH$_2$)
20	139	54% τ(N–N), 14% γ(C–CONHNH$_2$)
19	128	29% τ(N–N), 18% τ(C–N)
18	125	34% τ(C–N), 27% τ(N–N)
17	118	19% τ(C–N)
16	110	41% ν(HB)s b, 37% ν(HB)s a
15	96	20% τ(C–O)s, 20% τ(C4–C5), 12% γ(C–CONHNH$_2$)
14	88	38% τ(C–C)as, 24% τ(C–O)as
13	73	17% δ(im) 1 a, 17% δ(im) 1 b, 14% δ(CCC)as
12	66	19% ν(HB)s a, 19% ν(HB)s b, 12% δ(im) 1 b, 11% δ(im) 1 a
11	61	57% τ(C–C), 13% τ(im) 1 b, 11% τ(im) 3 a
10	60	59% τ(C–C), 11% τ(im) 3 a
9	54	34% τ(C–C)as
8	43	31% τ(C–C), 16% τ(C–C)s, 16% τ(im) 3 a, 14% τ(im) 1 b
7	38	16% τ(im) 2 b, 15% τ(im) 1 b, 12% τ(C4–C5), 11% τ(C–C)
6	38	20% τ(im) 3 a, 18% τ(C–C), 17% τ(im) 1 a
5	28	21% ν(HB)as a, 19% ν(HB)as b, 11% ν(HB)s a, 10% τ(C4–C5)
4	20	25% τ(im) 1 a, 21% τ(im) 2 b, 13% ν(HB)as b
3	13	20% ν(HB)as a, 16% ν(HB)as b, 15% δ(CCC)as, 10% τ(im) 2 b
2	10	54% τ(C–C)s, 12% τ(im) 1 a
1	3	51% τ(C4–C5), 16% τ(C–C)s, 10% τ(C–O)s

[a] Anharmonic wavenumbers (ṽ) in cm^{-1}; *ν*, bond stretching; *δ*, bending; *γ*, rocking; *τ*, torsion; w, wagging; tw, twisting; s, symmetric; as, asymmetric. For definition of coordinates see Tables S10, S11 and S13.

Table S24. Results of the normal coordinate analysis for SUC-INH-INH-SUC.[a]

Nº	ῡ	PED (≥10%)
180	3758	50% ν(O–H)s, 50% ν(O–H)as
179	3758	50% ν(O–H)s, 50% ν(O–H)as
178	3635	100% ν(N–H)
177	3635	100% ν(N–H)
176	3528	87% ν(NH_2)as, 12% ν(NH_2)s
175	3527	88% ν(NH_2)as, 11% ν(NH_2)s
174	3435	88% ν(NH_2)s, 11% ν(NH_2)as
173	3430	88% ν(NH_2)s, 12% ν(NH_2)as
172	3210	96% ν(C–H) 3
171	3210	96% ν(C–H) 3
170	3189	80% ν(C–H) 4, 19% ν(C–H) 2
169	3189	80% ν(C–H) 4, 19% ν(C–H) 2
167	3173	76% ν(C–H) 2, 20% ν(C–H) 4
168	3173	76% ν(C–H) 2, 20% ν(C–H) 4
166	3169	92% ν(C–H) 1
165	3169	92% ν(C–H) 1
164	3096	97% ν(CH_2 as)as
163	3096	97% ν(CH_2 as)as
162	3078	92% ν(CH_2 as)s
161	3078	92% ν(CH_2 as)s
160	3061	53% ν(CH_2 s)as, 20% ν(O–H)s, 20% ν(O–H)as
159	3061	67% ν(CH_2 s)as, 12% ν(O–H)s, 12% ν(O–H)as
158	3055	37% ν(CH_2 s)as, 26% ν(O–H)as, 26% ν(O–H)s
157	3053	34% ν(CH_2 s)s, 22% ν(O–H)as, 22% ν(O–H)s, 19% ν(CH_2 s)as
156	3051	86% ν(CH_2 s)s
155	3051	60% ν(CH_2 s)s, 17% ν(O–H)s, 17% ν(O–H)as
154	1808	46% ν(C=O)s, 39% ν(C=O)as
153	1808	46% ν(C=O)s, 39% ν(C=O)as
152	1761	44% ν(C=O)as, 35% ν(C=O)s
151	1761	44% ν(C=O)as, 35% ν(C=O)s
150	1730	53% δ(NH_2), 36% ν(C=O)
149	1722	84% δ(NH_2)
148	1713	43% δ(NH_2), 43% ν(C=O)
147	1705	71% ν(C=O), 10% δ(NH_2)
146	1644	65% ν_{Ring} 3, 23% δ(C–H) 2
145	1643	65% ν_{Ring} 3, 22% δ(C–H) 2
144	1596	72% ν_{Ring} 2
143	1596	73% ν_{Ring} 2
142	1534	31% δ(C–H) 4, 30% δ(N–H), 16% ν_{Ring} 6, 10% ν(N–C)
141	1533	34% δ(C–H) 4, 26% δ(N–H), 18% ν_{Ring} 6
140	1515	35% δ(N–H), 33% δ(C–H) 4, 15% ν_{Ring} 6, 10% ν(N–C)
139	1514	38% δ(N–H), 30% δ(C–H) 4, 14% ν_{Ring} 6, 12% ν(N–C)
138	1489	27% δ(COH)s, 27% δ(COH)as
137	1489	27% δ(COH)s, 27% δ(COH)as
136	1467	72% δ(CH)as, 20% δ(CH_2)s
135	1467	71% δ(CH)as, 20% δ(CH_2)s
134	1462	67% δ(CH_2)s, 23% δ(CH)as
133	1462	66% δ(CH_2)s, 23% δ(CH)as
132	1443	44% δ(C–H) 3, 31% ν_{Ring} 5
131	1443	44% δ(C–H) 3, 31% ν_{Ring} 5
129	1419	48% w(CH_2)s, 10% δ(CH_2)s
130	1419	48% w(CH_2)s, 10% δ(CH_2)s
128	1364	49% w(NH_2), 13% w(CH_2)as
127	1364	46% w(NH_2), 15% w(CH_2)as
126	1364	25% w(NH_2), 21% w(CH_2)as, 11% δ(COH)s, 11% ν(C–O)as
125	1364	71% w(NH_2)
124	1352	76% δ(C–H) 1, 12% ν_{Ring} 2
123	1352	76% δ(C–H) 1, 12% ν_{Ring} 2
122	1316	26% δ(N–H), 22% ν(N–C), 13% ν(C–C), 12% ν(N–N)

121	1314	25% δ(N–H), 23% ν(N–C), 13% ν(C–C), 12% ν(N–N)
120	1307	67% tw(CH_2)s
119	1307	67% tw(CH_2)s
118	1292	20% δ(COH)as, 18% tw(CH_2)s, 16% w(CH_2)as, 10% ν(C–O)s
117	1292	20% δ(COH)as, 18% tw(CH_2)s, 16% w(CH_2)as, 10% ν(C–O)s
116	1280	74% ν_{Ring} 4
115	1280	74% ν_{Ring} 4
114	1246	57% δ(C–H) 2, 24% ν_{Ring} 3
113	1246	58% δ(C–H) 2, 24% ν_{Ring} 3
112	1229	29% w(CH_2)as, 28% ν(C–O)s, 10% ν(C–O)as
111	1229	29% w(CH_2)as, 28% ν(C–O)s, 10% ν(C–O)as
110	1215	26% ν(N–N), 18% ν(C–C), 17% γ(NH_2), 12% ν_{Ring} 1, 10% δ_{Ring} 1
109	1213	27% ν(N–N), 19% ν(C–C), 14% γ(NH_2), 12% ν_{Ring} 1, 11% δ_{Ring} 1
107	1186	82% tw(CH_2)as
108	1186	82% tw(CH_2)as
106	1143	29% ν(C–O)as, 16% ω(CH_2)as, 13% ν(C–O)s, 11% δ(COH)as
105	1143	29% ν(C–O)as, 16% ω(CH_2)as, 13% ν(C–O)s, 11% δ(COH)as
104	1125	19% ν(N–C), 17% γ(NH_2), 13% ν_{Ring} 5, 12% δ(C–H) 3
103	1120	24% ν_{Ring} 5, 22% δ(C–H) 3, 12% ν(N–C), 12% γ(NH_2)
102	1112	33% ν_{Ring} 5, 31% δ(C–H) 3
101	1110	22% ν_{Ring} 5, 21% δ(C–H) 3, 14% ν(N–C), 12% γ(NH_2)
100	1089	35% ν_{Ring} 6, 20% δ_{Ring} 1, 16% δ(C–H) 4, 11% ν_{Ring} 1
99	1088	34% ν_{Ring} 6, 20% δ_{Ring} 1, 16% δ(C–H) 4, 11% ν_{Ring} 1
98	1083	67% ν(C–C)
97	1083	67% ν(C–C)
96	1044	68% γ(CH_2)s, 27% γ(C=O)as
95	1044	68% γ(CH_2)s, 27% γ(C=O)as
94	1031	26% τ(im) 3 ca, 17% τ(C–O)s, 17% τ(C–O)as, 14% τ(im) 3 bd
93	1031	26% τ(im) 3 bd, 17% τ(C–O)as, 17% τ(C–O)s, 14% τ(im) 3 ca
92	1028	48% ν_{Ring} 1, 47% δ_{Ring} 1
91	1028	48% ν_{Ring} 1, 46% δ_{Ring} 1
90	1021	42% γ(C–H) 4, 22% γ(C–H) 1, 19% γ(C–H) 2
89	1021	42% γ(C–H) 4, 22% γ(C–H) 1, 19% γ(C–H) 2
88	993	43% γ(C–H) 3, 38% γ(C–H) 2
87	993	43% γ(C–H) 3, 38% γ(C–H) 2
86	961	43% ν(N–N), 38% γ(NH_2)
85	954	41% ν(N–N), 39% γ(NH_2)
84	930	44% ν(C–C)s, 21% δ(CCC)s
83	930	44% ν(C–C)s, 21% δ(CCC)s
82	901	21% δ(C=O), 17% δ(CNN)
81	898	17% δ(C=O), 15% δ(CNN), 11% γ(NH_2)
80	893	50% γ(C–H) 1, 23% γ(C–H) 3, 12% γ(C–H) 2
79	893	46% γ(C–H) 1, 20% γ(C–H) 3
78	880	56% ν(C–C)as, 24% ν(C–O)as
77	880	56% ν(C–C)as, 24% ν(C–O)as
76	868	22% γ(C–H) 4, 18% γ(C=O), 17% γ(C–H) 3, 16% γ(C–$CONHNH_2$), 13% γ(C–H) 2
75	867	22% γ(C–H) 4, 17% γ(C=O), 17% γ(C–H) 3, 16% γ(C–$CONHNH_2$), 13% γ(C–H) 2
74	809	46% γ(CH_2)as, 35% γ(C=O)s, 14% tw(CH_2)as
73	809	46% γ(CH_2)as, 35% γ(C=O)s, 14% tw(CH_2)as
72	776	37% τ_{Ring} 1, 34% γ(C=O)
71	775	38% τ_{Ring} 1, 34% γ(C=O)
70	726	64% τ_{Ring} 1, 29% γ(C=O)
69	725	63% τ_{Ring} 1, 29% γ(C=O)
68	703	54% δ_{Ring} 3, 13% δ(C=O)
67	702	54% δ_{Ring} 3, 13% δ(C=O)
66	683	37% δ(C=O)s, 28% δ_{Ring} 2, 10% ν(C–O)s
65	683	37% δ(C=O)s, 28% δ_{Ring} 2, 10% ν(C–O)s
64	676	59% δ_{Ring} 2, 19% δ(C=O)s
63	676	59% δ_{Ring} 2, 19% δ(C=O)s
62	653	28% τ(C–O)s, 26% τ(C–O)as, 23% γ(C=O)as
61	653	28% τ(C–O)s, 26% τ(C–O)as, 23% γ(C=O)as
60	612	64% δ(C=O)as, 11% ν(C–C)as
59	612	64% δ(C=O)as, 11% ν(C–C)as

58	580	24% γ(C=O)as, 17% δ(C=O)as, 17% γ(C=O)s, 16% γ(CH_2)as
57	580	24% γ(C=O)as, 17% δ(C=O)as, 17% γ(C=O)s, 16% γ(CH_2)as
56	538	44% δ(C=O)as, 13% γ(C=O)s, 13% γ(CH_2)as, 11% δ(CCC)as
55	538	44% δ(C=O)as, 13% γ(C=O)s, 13% γ(CH_2)as, 11% δ(CCC)as
54	535	24% τ_{Ring} 2, 21% γ(N–H), 17% γ(C–$CONHNH_2$)
53	534	25% τ_{Ring} 2, 18% γ(C–$CONHNH_2$), 17% γ(N–H), 10% δ(CCO)
52	512	17% γ(C=O)as, 16% τ(C–O)s, 16% τ(C–O)as, 14% δ(C=O)as
51	512	18% γ(C=O)as, 16% τ(C–O)s, 16% τ(C–O)as, 14% δ(C=O)as
50	504	64% γ(N–H)
49	504	59% γ(N–H)
48	425	34% τ_{Ring} 2, 16% δ(C=O)
47	423	34% τ_{Ring} 2, 16% δ(C=O)
45	396	96% τ_{Ring} 3
46	396	96% τ_{Ring} 3
44	384	51% δ(C=O)s, 15% ν(C–C)
43	384	52% δ(C=O)s, 15% ν(C–C)
42	357	30% ν(C–C), 20% δ_{Ring} 3, 11% δ(C=O)s, 11% δ(CNN)
41	357	29% ν(C–C), 20% δ_{Ring} 3, 14% δ(CNN), 10% δ(C=O)s
40	318	24% δ(CNN), 17% τ(N–N), 14% δ(C=O), 12% δ(CCO)
39	312	21% δ(CNN), 12% δ(C=O), 11% τ(N–N), 11% γ(N–H), 10% δ(CCO), 10% δ(im) 1 ab
38	292	19% τ(N–N), 16% w(C–$CONHNH_2$), 14% τ(im) 3 ab, 12% δ(CNN), 10% τ(C–N)
37	289	24% τ(N–N), 18% τ(C–N), 18% w(C–$CONHNH_2$)
36	261	48% δ(CCC)s, 23% δ(C=O)s, 17% ν(C–C)s
35	261	48% δ(CCC)s, 23% δ(C=O)s, 17% ν(C–C)s
34	223	25% τ(C–N), 18% τ_{Ring} 2, 11% γ(C–$CONHNH_2$)
33	210	25% τ(N–N), 16% τ_{Ring} 2, 12% τ(C–N), 12% γ(C–$CONHNH_2$)
32	193	19% ν(HB)as ab, 19% w(C–$CONHNH_2$), 11% δ(CCO)
31	183	21% δ(CCC)as, 12% δ(im) 1 ca, 12% δ(im) 1 bd
30	173	30% δ(CCC)as
29	166	17% w(C–$CONHNH_2$), 14% τ(C–N), 14% δ(CCO), 12% ν(HB)s ab, 12% δ(CCC)as
28	144	17% γ(C–$CONHNH_2$), 16% ν(HB)s ab, 14% τ(C–N)
27	133	31% τ(C–N), 19% γ(C–$CONHNH_2$)
26	111	29% ν(HB)s ab
25	101	20% ν(HB)as ab
24	96	12% ν(HB)s ca, 12% ν(HB)s bd, 12% τ(C4–C5)
23	93	24% ν(HB)s bd, 24% ν(HB)s ca, 12% τ(C–C)as
22	92	9% ν(HB)s ca, 9% ν(HB)s bd, 9% ν(HB)as ab
21	88	17% δ(im) 1 ca, 17% δ(im) 1 bd, 10% ν(HB)s ca, 10% ν(HB)s bd
20	81	51% τ(C–C)as, 12% ν(HB)s ab
19	78	24% τ(C–C)as
18	70	41% τ(C–C)as
17	70	22% τ(C–C)as, 17% τ(C4–C5), 14% τ(C–C)
16	61	25% τ(C–C), 17% τ(N–N)
15	54	21% τ(im) 1 bd, 21% τ(im) 1 ca, 13% τ(C–C), 11% τ(im) 3 bd, 11% τ(im) 3 ca
14	53	15% τ(im) 1 ca, 15% τ(im) 1 bd, 13% τ(N–N), 13% τ(im) 3 ca, 12% τ(im) 3 bd, 11% τ(C–C)s
13	42	32% τ(C4–C5)
12	39	25% τ(C4–C5), 11% τ(im) 1 bd, 11% τ(im) 1 ca
11	33	18% τ(C–C), 18% ν(HB)as bd, 17% ν(HB)as ca
10	31	30% τ(C–C), 17% τ(C–C)s, 13% τ(N–N), 12% δ(im) 1 ab
9	25	24% τ(C–C), 12% τ(C–C)s
8	22	23% ν(HB)as ca, 23% ν(HB)as bd
7	18	32% τ(im) 3 ab, 23% δ(im) 1 ab, 13% τ(C–N)
6	13	21% τ(im) 1 ab, 15% τ(C4–C5)
5	10	56% τ(C–C)s
4	9	64% τ(C–C)s, 15% τ(C4–C5)
3	9	31% τ(C4–C5), 17% τ(im) 2 bd, 16% τ(im) 2 ca
2	5	38% τ(im) 1 ab, 28% τ(im) 2 ab
1	3	41% τ(im) 2 ab, 11% τ(N–N)

[a] Anharmonic wavenumbers (ṽ) in cm^{-1}; *ν*, bond stretching; *δ*, bending; *γ*, rocking; *τ*, torsion; w, wagging; tw, twisting; s, symmetric; as, asymmetric. For definition of coordinates see Tables S10, S11 and S14.

Table S25. Results of the normal coordinate analysis for INH-INH.[a]

Nº	ῡ	PED (≥10%)
96	3500	96% ν(NH_2)as
95	3500	97% ν(NH_2)as
94	3446	78% ν(NH_2)s, 19% ν(N–H)
93	3439	95% ν(NH_2)s
92	3424	98% ν(N–H)
91	3408	80% ν(N–H), 19% ν(NH_2)s
90	3205	97% ν(C–H) 3
89	3205	97% ν(C–H) 3
88	3190	95% ν(C–H) 4
87	3190	95% ν(C–H) 4
86	3157	88% ν(C–H) 1
85	3157	88% ν(C–H) 1
84	3152	86% ν(C–H) 2
83	3152	86% ν(C–H) 2
82	1726	72% ν(C=O), 10% δ(NH_2)
81	1725	70% ν(C=O), 13% δ(NH_2)
80	1698	85% δ(NH_2)
79	1695	84% δ(NH_2), 11% ν(C=O)
78	1632	63% ν_{Ring} 3, 22% δ(C–H) 2
77	1632	63% ν_{Ring} 3, 22% δ(C–H) 2
76	1597	73% ν_{Ring} 2
75	1597	74% ν_{Ring} 2
74	1558	59% δ(N–H), 11% ν(N–C)
73	1546	63% δ(N–H), 12% ν(N–C)
72	1518	60% δ(C–H) 4, 30% ν_{Ring} 6
71	1518	57% δ(C–H) 4, 28% ν_{Ring} 6
70	1438	50% δ(C–H) 3, 33% ν_{Ring} 5
69	1438	50% δ(C–H) 3, 33% ν_{Ring} 5
68	1353	58% δ(C–H) 1, 17% w(NH_2)
67	1352	77% δ(C–H) 1, 11% ν_{Ring} 2
66	1349	57% w(NH_2), 20% δ(C–H) 1
65	1343	80% w(NH_2)
64	1325	22% ν(N–C), 20% w(NH_2), 16% ν(C–C)
63	1320	24% ν(N–C), 18% ν(C–C), 12% w(NH_2)
62	1276	77% ν_{Ring} 4
61	1276	76% ν_{Ring} 4
60	1244	58% δ(C–H) 2, 25% ν_{Ring} 3
59	1243	58% δ(C–H) 2, 25% ν_{Ring} 3
58	1207	32% γ(NH_2), 23% ν(N–N), 12% ν(C–C)
57	1206	25% γ(NH_2), 23% ν(N–N), 15% ν(C–C), 11% ν_{Ring} 1
56	1141	28% γ(NH_2), 23% ν(N–C)
55	1130	32% γ(NH_2), 21% ν(N–C)
54	1109	44% ν_{Ring} 5, 39% δ(C–H) 3
53	1108	41% ν_{Ring} 5, 36% δ(C–H) 3
52	1089	35% ν_{Ring} 6, 20% δ_{Ring} 1, 19% δ(C–H) 4, 11% δ(C–H) 2
51	1089	34% ν_{Ring} 6, 20% δ_{Ring} 1, 18% δ(C–H) 4, 10% δ(C–H) 2
50	1011	46% ν_{Ring} 1, 44% δ_{Ring} 1
49	1011	49% ν_{Ring} 1, 43% δ_{Ring} 1
48	1006	52% γ(C–H) 2, 28% γ(C–H) 3, 13% γ(C–H) 4
47	1006	52% γ(C–H) 2, 28% γ(C–H) 3, 13% γ(C–H) 4
45	999	46% ν(N–N), 21% γ(NH_2)
46	996	50% ν(N–N), 22% γ(NH_2)
44	983	41% γ(C–H) 4, 31% γ(C–H) 1, 16% γ(C–H) 3
43	982	40% γ(C–H) 4, 30% γ(C–H) 1, 16% γ(C–H) 3
42	912	21% δ(C=O), 20% δ(CNN)
41	903	22% δ(C=O), 19% δ(CNN)
40	895	55% γ(C–H) 1, 18% γ(C–H) 4, 18% γ(C–H) 3
39	894	55% γ(C–H) 1, 18% γ(C–H) 4, 17% γ(C–H) 3
38	858	28% γ(C–H) 3, 21% γ(C=O), 19% γ(C–H) 2, 16% γ(C–$CONHNH_2$)

37	857	28% γ(C–H) 3, 20% γ(C=O), 19% γ(C–H) 2, 16% γ(C–$CONHNH_2$)
36	783	23% γ(N–H), 15% τ(im) 2, 14% γ(C=O)
35	771	41% τ_{Ring} 1, 29% γ(C=O)
34	761	50% τ_{Ring} 1, 17% γ(N–H)
33	730	49% τ_{Ring} 1, 24% τ(im) 3, 15% γ(C=O)
32	718	50% τ_{Ring} 1, 37% γ(C=O)
31	710	31% τ(im) 3, 18% γ(C=O), 16% τ_{Ring} 1
30	689	53% δ_{Ring} 3, 15% δ(C=O)
29	688	44% δ_{Ring} 3, 15% δ(C=O)
28	680	85% δ_{Ring} 2
27	679	77% δ_{Ring} 2
26	519	25% τ_{Ring} 2, 16% δ(CCO), 16% γ(C–$CONHNH_2$)
25	516	27% τ_{Ring} 2, 17% γ(C–$CONHNH_2$), 16% δ(CCO)
24	440	35% τ_{Ring} 2, 14% δ(C=O)
23	438	32% τ_{Ring} 2, 16% δ(C=O)
22	385	98% τ_{Ring} 3
21	385	98% τ_{Ring} 3
20	359	29% δ(CNN), 22% ν(C–C), 18% δ_{Ring} 3
19	351	34% ν(C–C), 28% δ_{Ring} 3, 13% δ(CNN)
18	333	18% δ(C=O), 15% δ(CCO), 14% w(C–$CONHNH_2$), 12% ν(C–C), 11% δ_{Ring} 3, 11% δ(CNN)
17	329	35% δ(CNN), 19% δ(C=O), 15% w(C–$CONHNH_2$), 12% δ(CCO)
16	305	82% τ(N–N), 15% γ(N–H)
15	270	65% τ(N–N), 31% τ(im) 1
14	251	44% τ(C–N), 16% w(C–$CONHNH_2$), 16% τ_{Ring} 2
13	249	47% τ(C–N), 20% τ_{Ring} 2, 13% w(C–$CONHNH_2$)
12	187	22% ν(HB)s, 18% δ(im), 14% w(C–$CONHNH_2$), 11% τ_{Ring} 2, 11% τ(C–N)
11	164	35% w(C–$CONHNH_2$), 23% δ(CCO)
10	136	33% γ(C–$CONHNH_2$), 17% τ_{Ring} 2, 12% γ(N–H), 10% ν(HB)as
9	133	49% ν(HB)s, 14% w(C–$CONHNH_2$), 10% δ(CCO)
8	125	29% γ(C–$CONHNH_2$), 23% τ(C–N), 13% τ_{Ring} 2
7	66	37% γ(N–H), 30% τ(C–C), 14% τ(im) 3
6	61	64% τ(C–C), 13% γ(N–H)
5	44	42% δ(im), 11% τ(C–N)
4	28	54% ν(HB)as, 15% τ(C–N)
3	28	50% τ(C–C), 26% γ(N–H), 12% τ(im) 3
2	21	28% τ(C–C), 24% τ(im) 2, 21% γ(N–H)
1	10	44% τ(im) 1, 32% τ(im) 2, 11% τ(C–N), 10% γ(N–H)

[a] Anharmonic wavenumbers (ῡ) in cm^{-1}; *ν*, bond stretching; *δ*, bending; *γ*, rocking; *τ*, torsion; w, wagging; s, symmetric; as, asymmetric. For definition of coordinates see Tables S10, S11 and S15.

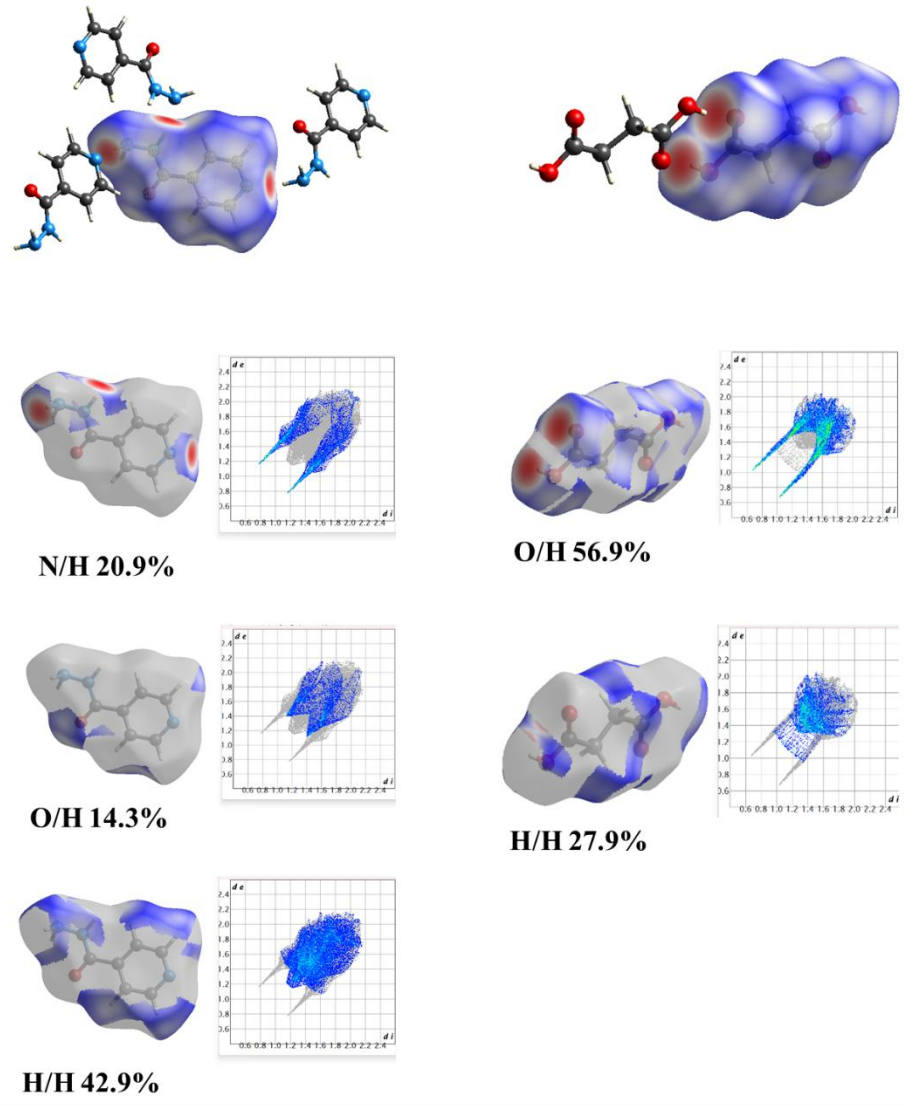

Fig. S1. Hirshfeld surfaces for the INH molecule in the INH (polymorph 1) crystal [21] and SUC molecule in the SUC (polymorph β) crystal [55] and the 2D fingerprint plots for the different relevant contacts in the two crystals.

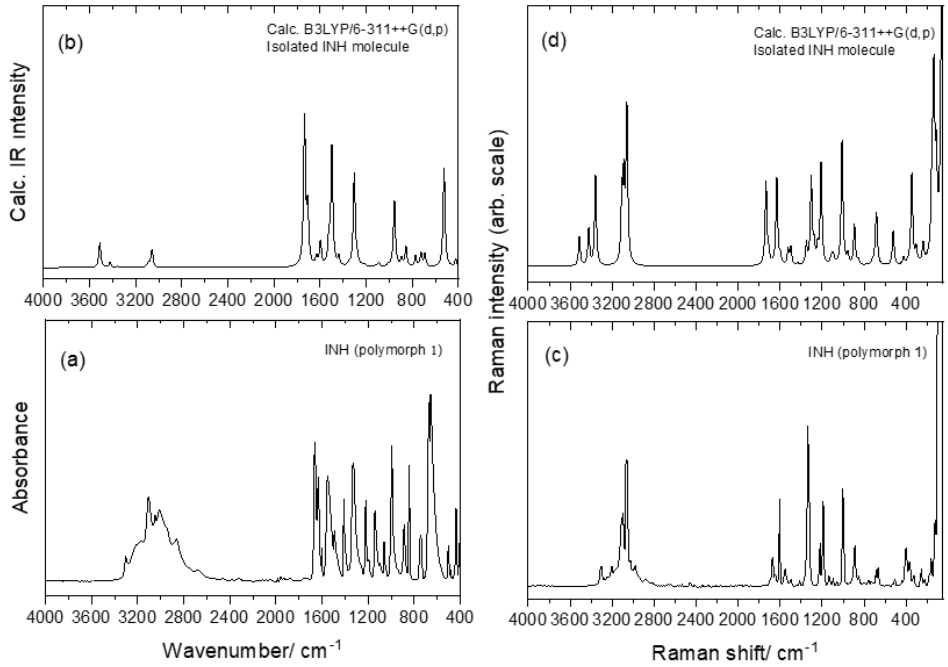

Fig. S2. Selected regions of the experimental room temperature infrared (a) and Raman (c) spectra of the INH crystal (polymorph 1), and B3LYP/6-311G++(d,p) calculated harmonic infrared (b) and Raman (d) spectra for the isolated INH molecule. In the simulated spectra bands were represented by Lorentzian functions centred at the calculated maxima and with FWHW = 20 cm^{-1}; above 2800 cm^{-1} wavenumbers were scaled by 0.97.

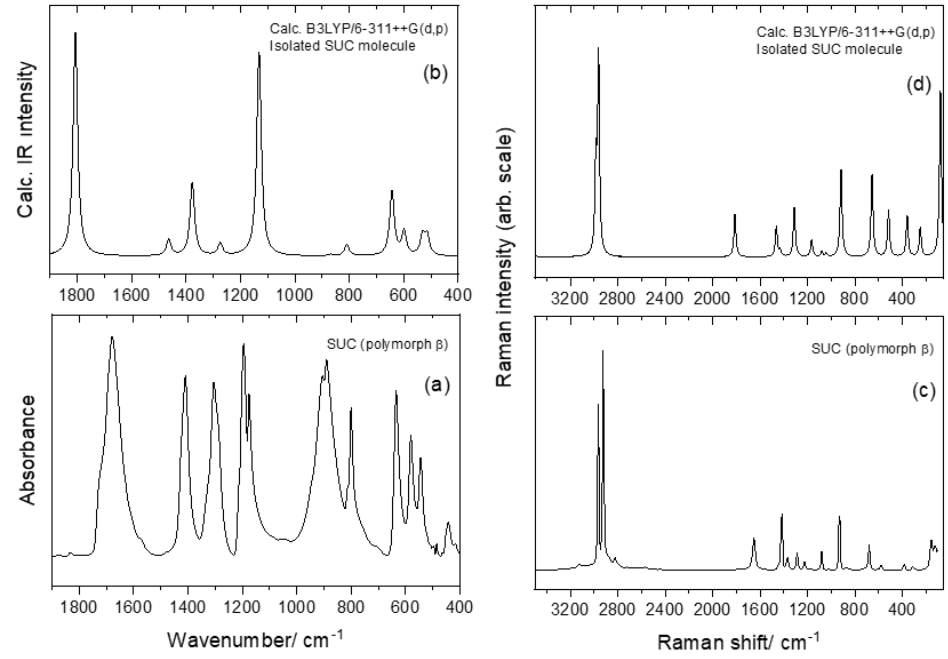

Fig. S3. Selected regions of the experimental room temperature infrared (a) and Raman (c) spectra of the SUC crystal (polymorph β), and B3LYP/6-311G++(d,p) calculated harmonic infrared (b) and Raman (d) spectra for the isolated SUC molecule. In the simulated spectra bands were represented by Lorentzian functions centred at the calculated maxima and with FWHW = 20 cm^{-1}; above 2800 cm^{-1} wavenumbers were scaled by 0.97.

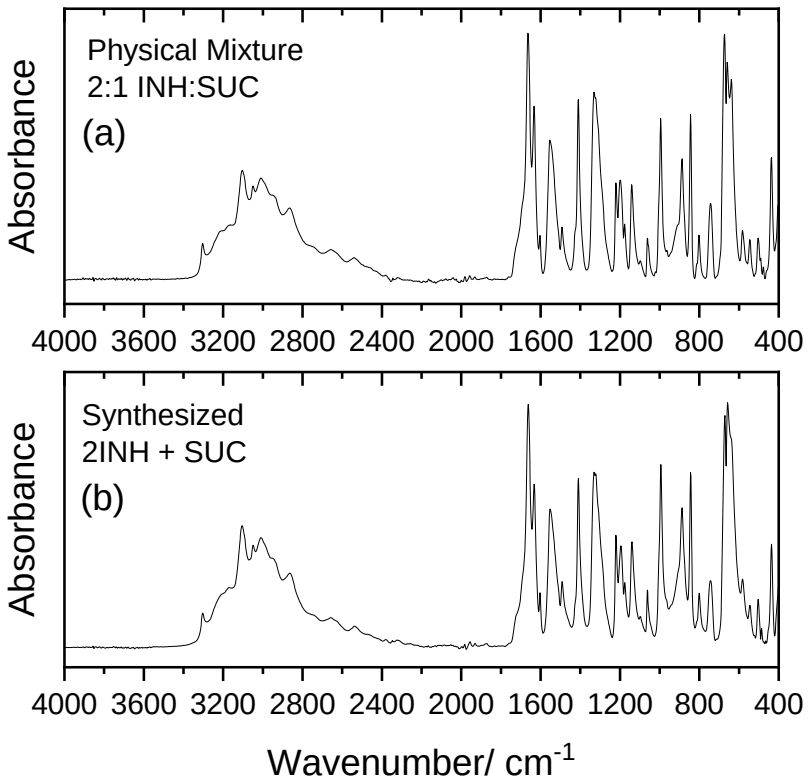

Fig. S4. Experimental room temperature IR spectrum of the INH:SUC 2:1 mixture (a), and sum spectrum synthesized using the experimental room temperature IR spectra of INH and SUC in a proportion 2:1 (b).

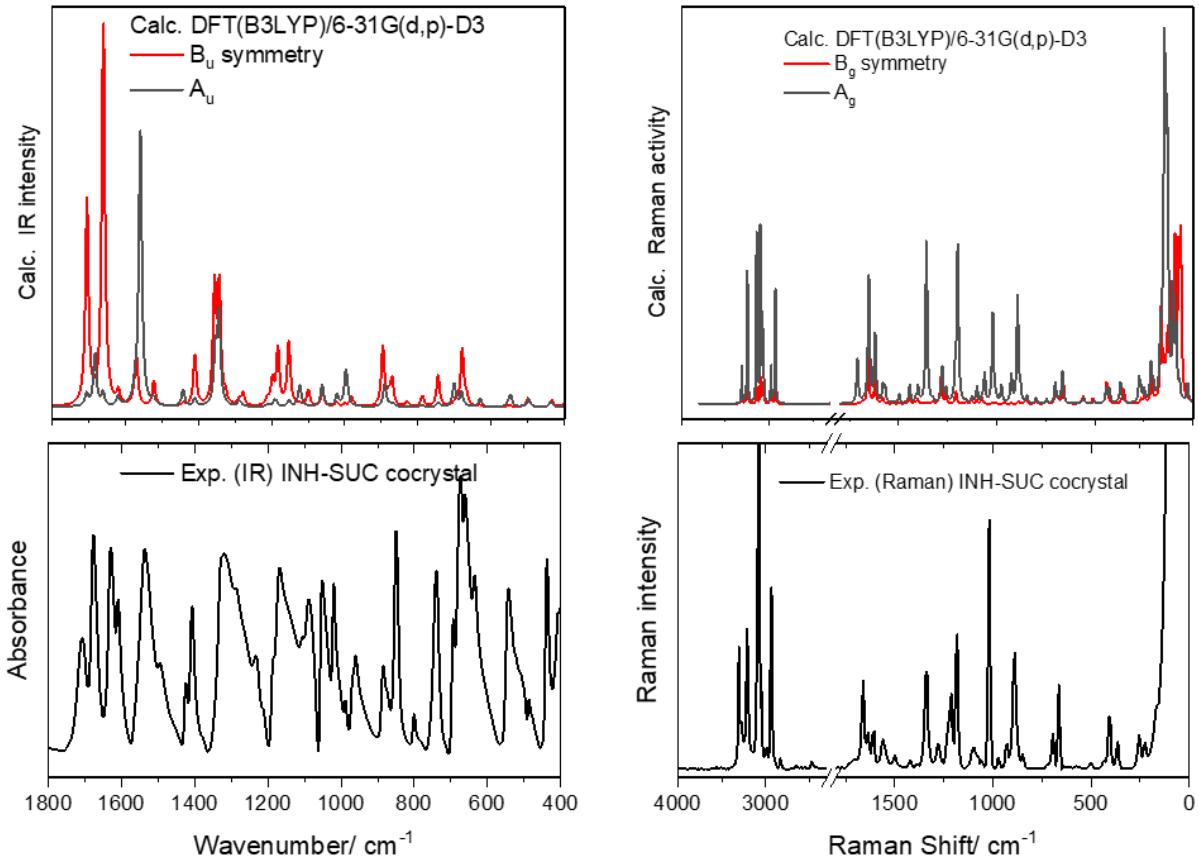

Fig. S5. Selected regions of the experimental room temperature infrared (left) and Raman (right) spectra of the INH-SUC cocrystal, and B3LYP/6-31G(d,p)-D3 full periodic vibrational calculations for the cocrystal. In the simulated spectra bands were represented by Lorentzian functions centred at the calculated maxima and with FWHW = 10 cm^{-1}; wavenumbers were scaled using scale factors of 0.95 and 0.97, above and below 1800 cm^{-1}, respectively.