Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Figure 1. Molecular packing (double unit cell) of sarcosine

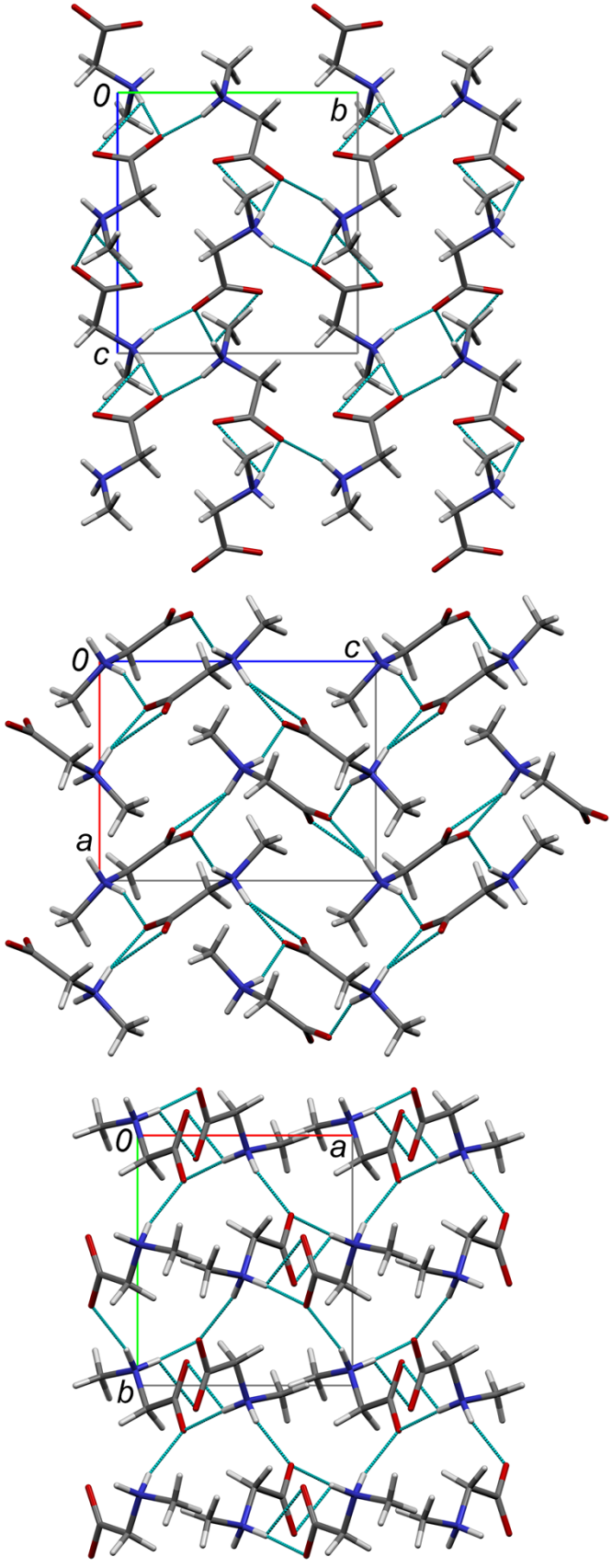

Figure 2. Molecular packing (double unit cell) of betaine

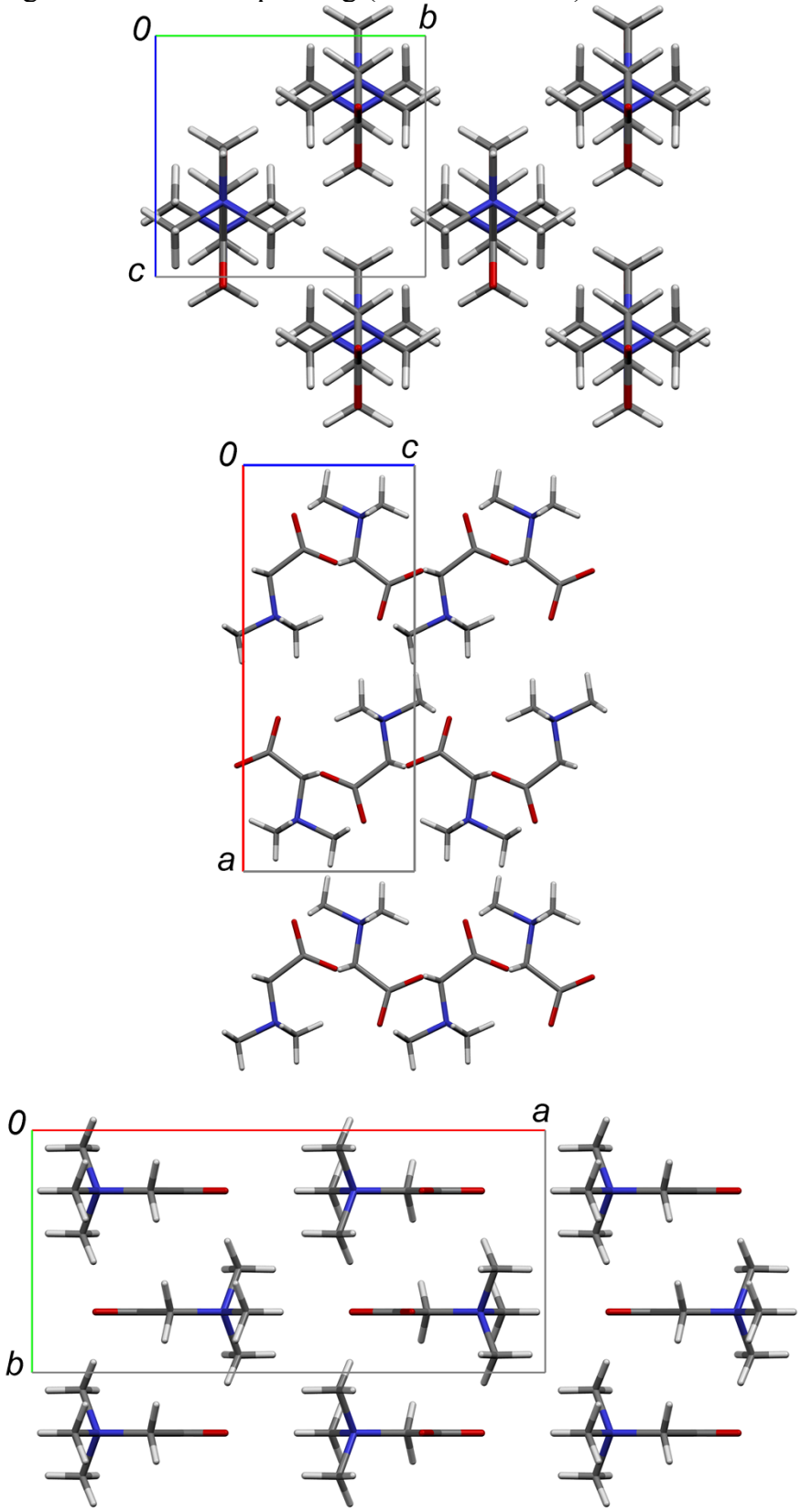

Figure 3. Changes in distances of C1-O1 bond (black circles) and C1-O2 bond (red rhombs) with variations in temperature in sarcosine. All curves are guides to the eye.

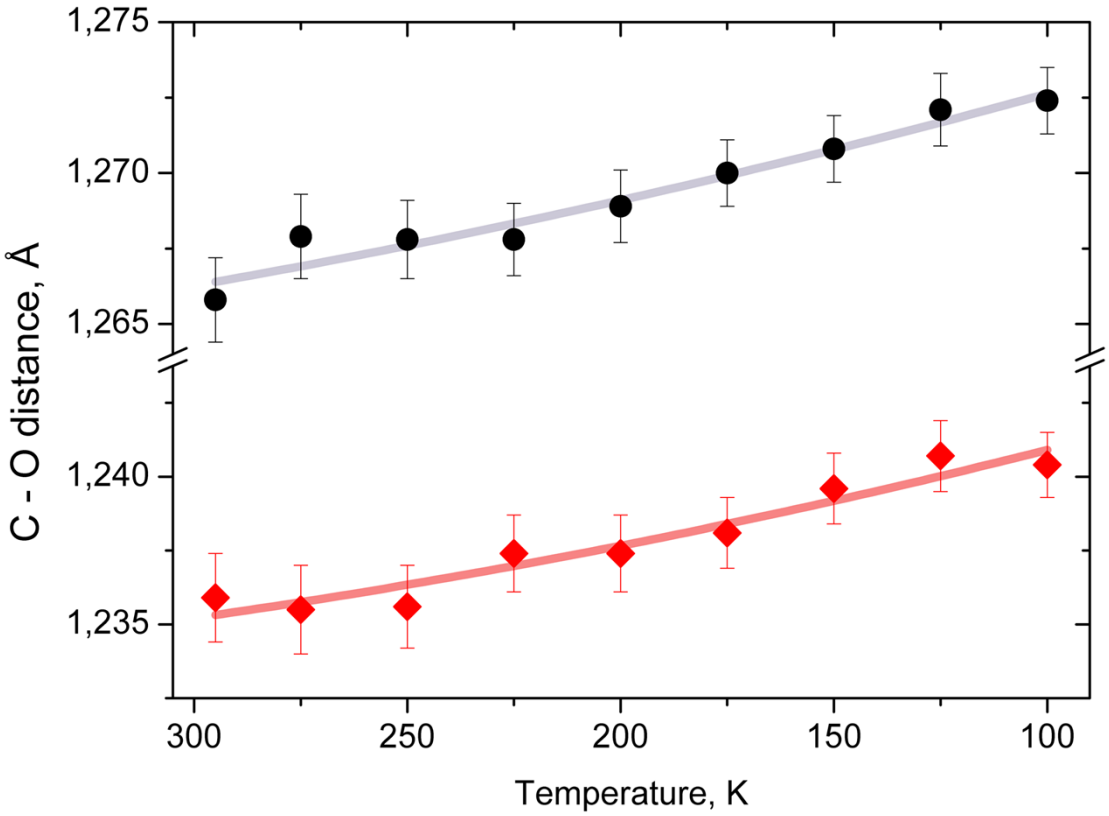

Figure 4. Non-polarized Raman spectra of sarcosine on cooling from ambient temperature down to 5 K. Red arrow shows the region of non-linear increasing of intensity of bands at 2432 cm^{-1} corresponding to the self-trapped state (see text)

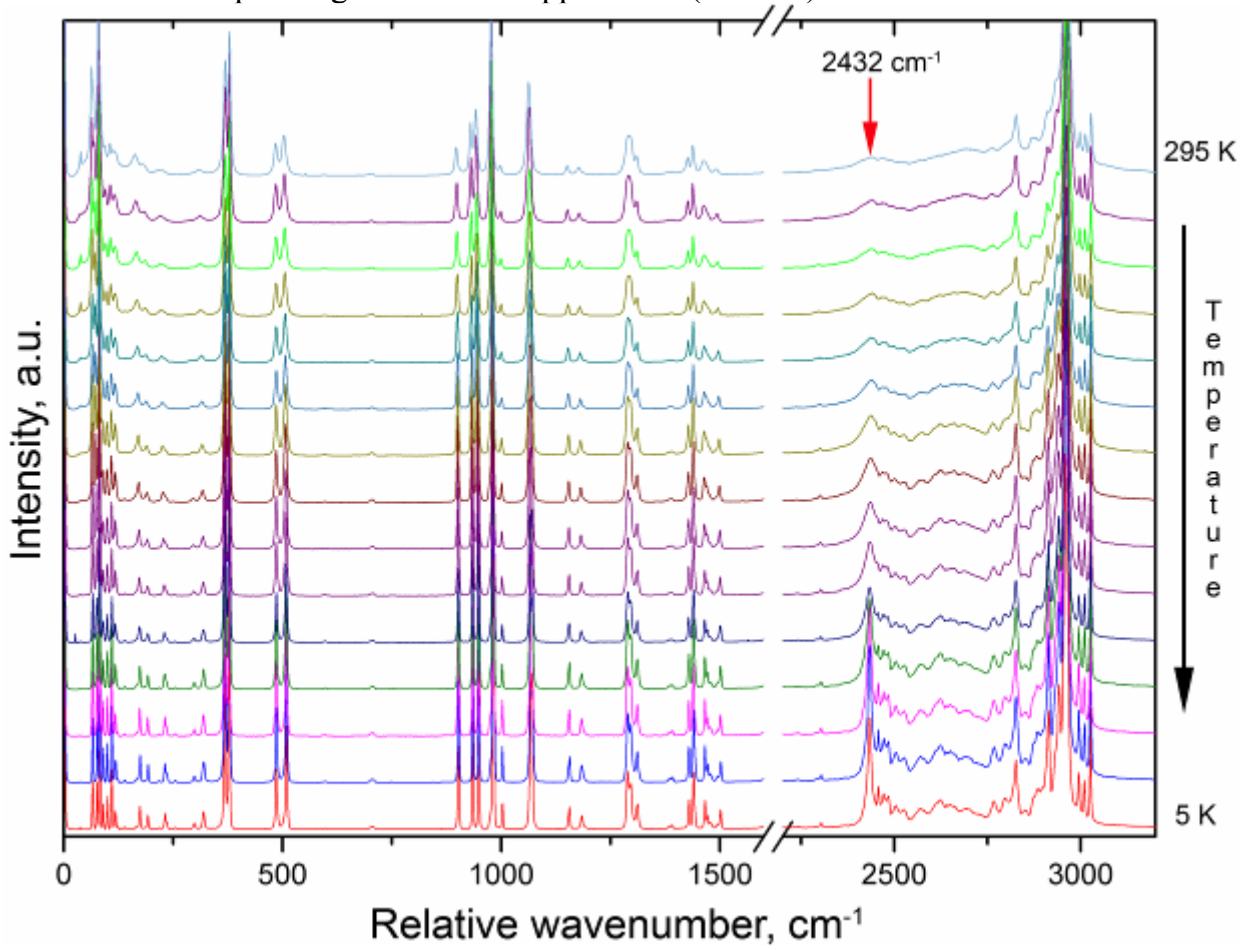

Figure 5. Normalized integral intensity of the band at 2432 cm^{-1}.

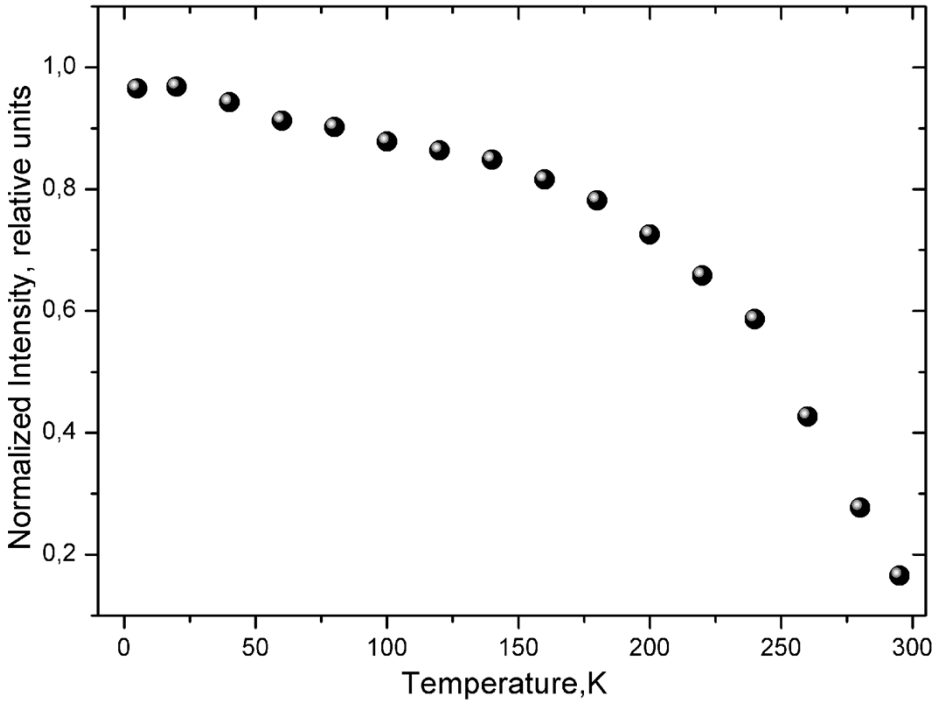

Table 1. Crystal data, data collection and structure refinement parameters for sarcosine at different temperatures†

T/K	295	275	250	225	200	175	150	125	100
a/Å	6.8141(5)	6.7970(5)	6.7737(5)	6.7518(4)	6.7285(4)	6.7099(4)	6.6925(4)	6.6768(4)	6.6605(4)
b/Å	7.9189(8)	7.9162(8)	7.9170(8)	7.9135(7)	7.9107(7)	7.9090(7)	7.9071(7)	7.9039(7)	7.9000(7)
c/Å	8.5887(6)	8.5951(6)	8.5989(5)	8.6022(5)	8.6056(5)	8.6098(5)	8.6136(5)	8.6154(5)	8.6170(5)
V/Å^3	463.45(7)	462.47(7)	461.14(6)	459.62(6)	458.05(6)	456.91(6)	455.82(6)	454.66(6)	453.41(6)
D_{calc}/g cm^{-3}	1.277	1.280	1.283	1.288	1.292	1.295	1.298	1.302	1.305
μ/mm^{-1}	0.107	0.107	0.107	0.107	0.108	0.108	0.108	0.109	0.109
No. of measured, independent, and observed [$I > 2\sigma(I)$] reflections	5047 1246 1077	5019 1246 1103	5017 1241 1081	5009 1238 1106	4986 1234 1121	4982 1232 1126	4956 1224 1128	4955 1221 1133	4931 1220 1141
θ_{min} (°)	3.50	3.50	3.50	3.50	3.50	3.50	3.50	3.50	3.50
θ_{max} (°)	29.16	29.15	29.16	29.17	29.19	29.16	29.15	29.17	29.16
R_{int}	0.0266	0.0279	0.0263	0.0271	0.0270	0.0251	0.0290	0.0275	0.0266
$R[F^2 > 2\sigma(F^2)]$	0.0330	0.0341	0.0314	0.0290	0.0291	0.0265	0.0254	0.0260	0.0252
$wR(F^2)$	0.0751	0.0747	0.0701	0.0636	0.0672	0.0617	0.0633	0.0605	0.0591
S	1.049	1.085	1.096	1.066	1.036	1.074	1.038	1.076	1.093
$\Delta\rho_{max}$/eÅ^{-3}	0.137	0.125	0.137	0.131	0.152	0.152	0.141	0.182	0.212
$\Delta\rho_{min}$/eÅ^{-3}	-0.201	-0.199	-0.220	-0.174	-0.190	-0.160	-0.157	-0.150	-0.183

†For all structures: chemical formula - $C_3H_7NO_2$, M_r = 89.10, orthorhombic, $P2_12_12_1$, Z = 4, Z' = 1, crystal size 0.30x0.22x0.10 mm, number of parameters - 62, range of h k l (-9 → 8; -10 → 10; -10 → 11). Experiments were carried out with Mo-K_α radiation using an IPDS-II Stoe diffractometer. H atoms treated by a mixture of independent and constrained refinement.

Table 2. Crystal data, data collection and structure refinement parameters for betaine at different temperatures†

T/K	295	275	250	225	200	175	150	125	100
a/Å	14.5652(16)	14.5609(16)	14.5561(15)	14.5514(15)	14.5449(14)	14.5416(13)	14.5379(13)	14.5340(13)	14.5314(13)
b/Å	6.8755(6)	6.8598(6)	6.8446(5)	6.8288(5)	6.8144(5)	6.8003(4)	6.7879(4)	6.7770(4)	6.7663(4)
c/Å	6.1405(5)	6.1290(5)	6.1179(5)	6.1066(5)	6.0963(4)	6.0871(4)	6.0772(4)	6.0668(4)	6.0577(4)
V/Å^3	614.93(10)	612.19(10)	609.53(9)	606.80(9)	604.23(8)	601.94(8)	599.71(8)	597.56(8)	595.62(8)
D_{calc}/g cm^{-3}	1.265	1.271	1.277	1.282	1.288	1.293	1.297	1.302	1.306
μ/mm^{-1}	0.097	0.097	0.098	0.098	0.099	0.099	0.099	0.100	0.100
No. of measured, independent, observed [$I > 2\sigma(I)$] reflections	5185 823 639	5419 856 659	5624 891 691	5582 884 708	5562 881 718	5547 877 738	5539 875 731	5501 873 733	5472 868 746
θ_{min} (°)	2.80	2.80	2.80	2.80	2.80	2.80	2.80	2.80	2.80
θ_{max} (°)	28.28	28.70	29.15	29.14	29.14	29.15	29.17	29.18	29.15
Range of									
h	-19 → 17	-17 → 19	-19 → 17	-19 → 17	-19 → 17	-18→ 19	-19 → 18	-19 → 18	-18→ 19
k	-9 → 9	-9 → 9	-9 → 9	-9 → 9	-9 → 9	-9 → 9	-9 → 9	-9 → 9	-9 → 9
l	-8 → 8	-8 → 8	-8 → 8	-8 → 8	-8 → 8	-8 → 8	-8 → 8	-8 → 8	-8 → 8
R_{int}	0.0309	0.0327	0.0325	0.0324	0.0334	0.0323	0.0356	0.0372	0.0370
$R[F^2 > 2\sigma(F^2)]$	0.0420	0.0416	0.0404	0.0410	0.0375	0.0365	0.0369	0.0359	0.0347
$wR(F^2)$	0.0969	0.0966	0.0956	0.0951	0.0886	0.0882	0.0893	0.0876	0.0839
S	1.051	1.055	1.076	1.077	1.083	1.101	1.083	1.079	1.102
$\Delta\rho_{max}$/eÅ^{-3}	0.143	0.133	0.164	0.138	0.155	0.165	0.140	0.160	0.188
$\Delta\rho_{min}$/eÅ^{-3}	-0.187	-0.254	-0.227	-0.254	-0.250	-0.241	-0.320	-0.290	-0.304

†For all structures: chemical formula - $C_5H_{11}NO_2$, M_r = 117.15, orthorhombic, Pnma, Z = 4, Z' =1, crystal size 0.45x0.40x0.10 mm, number of parameters - 63. Experiments were carried out with Mo-K_α radiation using an IPDS-II Stoe diffractometer. H atoms treated by independent refinement.